Observation of interdot energy transfer between InAs quantum dots

K. Nishibayashi,[1,a)] T. Kawazoe,[1] M. Ohtsu,[1] K. Akahane,[2] and N. Yamamoto[2]
[1]*Department of Electronics Engineering, University of Tokyo, 2-11-16, Yayoi, Bunkyo-ku Tokyo 113-8656, Japan*
[2]*National Institute of Information and Communications Technology, Koganei, Tokyo 1848795, Japan*

(Received 11 May 2008; accepted 21 May 2008; published online 28 July 2008)

We observed interdot excitation energy transfer between vertically aligned InAs quantum dots (QDs) separated by a 24-nm-thick spacer layer. This transfer was explained by resonant energy transfer via an optical near-field interaction between the first excited state of small QDs and the second excited states of large QDs. The excitation intensity dependence of the photoluminescence intensities showed that the energy transfer times were 75 ps at 15 K and 4 ns at 77 K. Our experimental results indicated that the III-V compound semiconductor QDs are appropriate for nanophotonic switching devices. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2945289]

A great deal of research interest exists in the excitation energy transfer between semiconductor quantum dots (QDs), and many investigations of interlayer excitation energy transfer have been conducted between self-assembled InAs QDs based on carrier transfer.[1–4] The electron tunneling process based on electron coupling between asymmetric InAs QDs has been reported to follow a semiclassical Wentzel–Kramers–Brillouin approximation,[2,3] and therefore, their time extends exponentially with increasing width of the spacer layer. In this case, the excitation energy transfer length between QDs is limited to several nanometers. Studies have also reported on carrier transfer processes based on thermal-assisted phenomena, such as the nonresonant multiphonon-assisted tunneling process (NPTP),[4] which is effective at high temperatures.

Kagan *et al.* reported excitation energy transfer between closely packed CdSe QDs and explained them via dipole-dipole interactions[5] corresponding to the Förster mechanism. Although the Förster mechanism is a valid model to explain the energy transfer between QDs, it is difficult to use for describing the dipole-forbidden transition[6] or for studying the effects of host materials and substrates for the QDs. Recently, we reported excitation energy transfer based on a transfer mechanism via an optical near-field (ONF) interaction.[7,8] In this model, the QDs are coupled by the virtual exciton polariton (i.e., an ONF), which involves the effects of the host and substrate as its effective mass. This model describes the observed optically forbidden energy transfer between CuCl QDs.[9] The ONF interaction is given by the Yukawa function[10] and the transfer length is in the range of several tens of nanometers. Using this feature, we demonstrated nanophotonic AND- and NOT-gate operations.[11–13] Here, we discuss the observation of fast energy transfer of 75 ps via an ONF between vertically stacked InAs/GaAs QDs with a 24-nm-thick spacer layer.

Our samples were grown using molecular beam epitaxy in Stranski–Krastanov (SK) mode on a (001)-oriented GaAs substrate. We prepared two samples: single-layer InAs QDs (SQDs) and double-layer QDs (DQDs). After deposition of a 200-nm-thick GaAs buffer layer, a 2 ML InAs layer was grown to fabricate self-assembled InAs QDs. For the SQD sample, a 24-nm-thick GaAs layer was deposited on the QDs as a cap layer. For the DQD sample, a second InAs QD layer was grown on the 24 nm spacer layer under the same growth conditions used for the first InAs layer. Finally, a 24-nm-thick GaAs cap layer was deposited on the second InAs QD layer. The size of the SQDs was estimated to be 14 nm in height and 32 nm in width using a transmission electron microscope image. For the DQD sample, the first and second layer QDs were estimated to be 14 nm in height and 32 nm in width, and 18 nm in height and 38 nm in width, respectively. We suspect that the larger size of the second layer QDs than that of the first layer QDs is due to the local stress.[14] In the following discussion, the QDs in the first and second layers are designated as QD-*A* and QD-*B*, respectively. The insets in Fig. 1 show schematic representa-

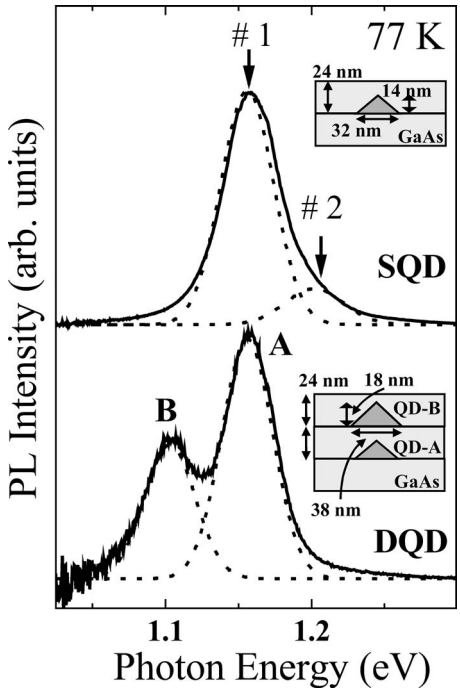

FIG. 1. PL spectra of the SQD (upper) and DQD (lower) samples at 77 K. The dashed curve shows two peaks fitted to the PL spectrum of the DQD sample. The insets show the schematic structures of the samples.

[a)]Author to whom correspondence should be addressed. Electronic mail: nishib@ee.t.u-tokyo.ac.jp.

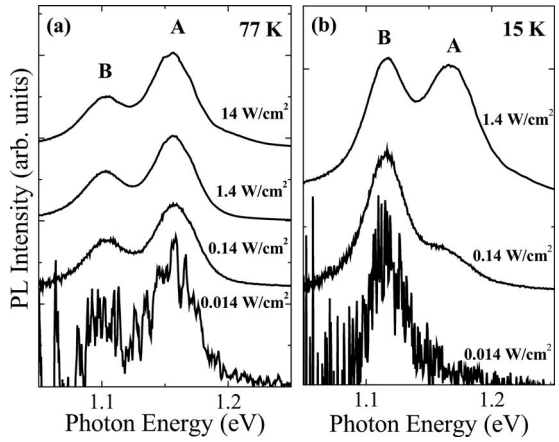

FIG. 2. Normalized PL spectra of the DQD sample with various excitation intensities at 77 (a) and 15 K (b).

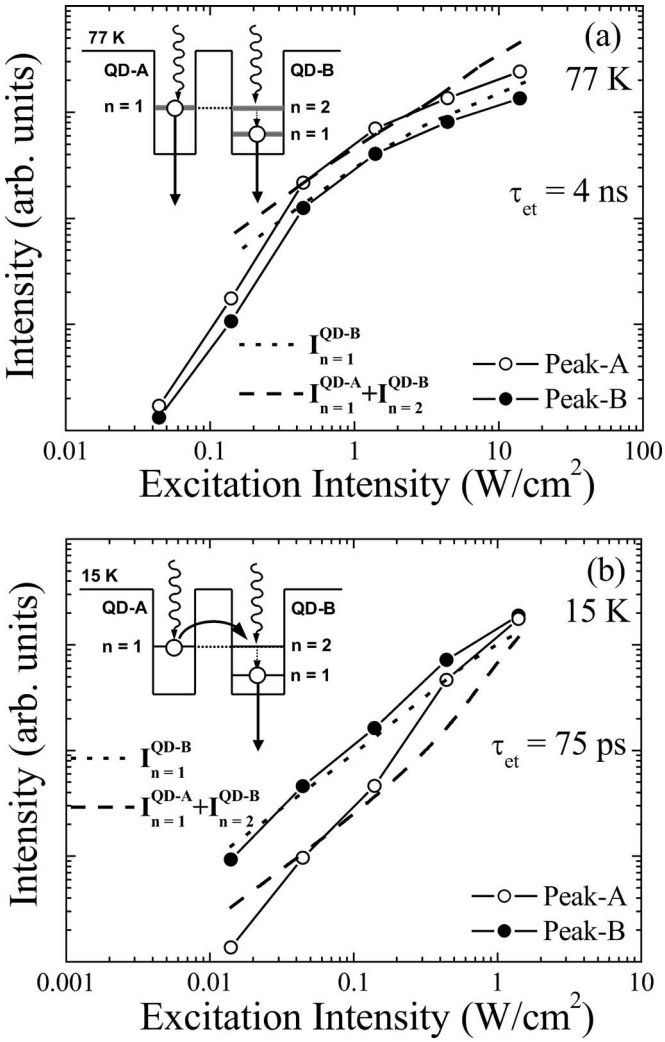

FIG. 3. Excitation intensity dependence of the integrated PL intensities of experimental results at 77 (a) and 15 K (b). The lines with open and closed circles show the integrated PL intensities of peaks *A* and *B*, respectively. The broken and dotted lines show the respective fitting results.

tions of the sample structure. The density of the QDs in the two samples was estimated to be $5\times10^9/\mathrm{cm}^2$. The average distance between each InAs QD was 340 nm, and thus interlayer carrier hopping[15,16] can be neglected in our discussion.

The photoluminescence (PL) spectra of the SQD and DQD samples at 77 K are shown in Fig. 1. We used second harmonic generation of a mode-locked Ti:sapphire laser as an excitation light source. The excitation photon energy was 2.754 eV with an excitation density of 14 $\mathrm{W/cm}^2$. In the PL spectrum of the SQD sample, a PL peak appeared at 1.156 eV (No. 1) corresponding to the lowest exciton level ($n=1$). The asymmetrical tail of the PL spectrum for the higher energy side was due to the emission of the second excited state ($n=2$) located at 1.202 eV (No. 2). By Gaussian fitting, the full width at half maximum of the PL peak and the energy difference between the $n=1$ and $n=2$ states were estimated as 35 and 46 meV, respectively.

For the DQD sample, two clearly separated peaks in the PL spectral peak appeared at 1.155 eV (*A*) and 1.103 eV (*B*). As the PL-peak energy at 1.155 eV corresponded to that of SQD samples, PL-peak *A* was due to emission of the lower layer QDs, QD-*A*, the sizes of which were the same as those of the SQD sample. The PL-peak *B* at 1.103 eV was due to PL of the upper layer QD, QD-*B*, which was larger than that of SQD.

Figure 2(a) shows the normalized PL spectra of the DQD sample under various excitation intensities at 77 K. Two PL peaks (*A* and *B*) are revealed. Figure 3(a) shows the excitation intensity dependence of the integrated PL intensities of the two PL bands. Both PL intensities increased nonlinearly in the region of excitation intensity <0.5 $\mathrm{W/cm}^2$, which can be explained by saturation of the thermally activated nonradiative relaxation path around QDs (Ref. 17) or around the QD interface. In the region of excitation intensity >0.5 $\mathrm{W/cm}^2$, the PL intensities increased linearly. The ratio of PL intensity for peaks *A* and *B* remained constant even with increasing excitation intensity, supporting the conclusion that QD-*A* and QD-*B* emissions are independent and the interaction between them is weak.

The PL spectra of the DQD sample at 15 K are shown in Fig. 2(b), which were different from those observed at 77 K. PL peaks *A* and *B* shifted to the higher energy side by 14 meV.[18] Peak *A* disappeared at the lowest excitation intensity (0.014 $\mathrm{W/cm}^2$), but appeared at 1.167 eV with increasing excitation intensity (0.14 $\mathrm{W/cm}^2$). At the highest excitation intensity (1.4 $\mathrm{W/cm}^2$), the PL intensities of the two peaks' heights became almost the same. Figure 3(b) shows the excitation intensity dependence of the integrated PL intensities around the two peaks at 15 K. The relative intensity of peak *A* increased more rapidly than that of peak *B* with increasing excitation intensity, indicating that effective excitation energy transfer occurs between QD-*A* and -*B*.

In our sample, electron tunneling by interlayer electron coupling[2] was negligible due to the spacer layer of 24 nm. The NPTP (Ref. 4) was also inconsistent with our results, as this process is ineffective at low temperatures. To explain our experimental results regarding the excitation intensity dependences at 15 and 77 K, we propose a model of vertical coupling via an ONF between QD-*A* and QD-*B*, as shown in the inset of Fig. 3(b). We assumed that the $n=1$ state of QD-*A* and the $n=2$ state of QD-*B* resonate with each other because the energy difference of 51 meV between the emission states of QD-*A* and QD-*B* is close to the energy difference of 46 meV between the first and second excited states of the SQD sample observed at 15 K. Within a few tens of picoseconds after excitation, photogenerated excitons or electron-hole pairs are captured in the QD-*A* and QD-*B* at 15 K [see

inset of Fig. 3(b)], and the excitons of the $n=1$ state of QD-*A* transfer to the $n=2$ state of QD-*B* at low excitation intensities. They relax to the $n=1$ state of QD-*B* with phonon emission within several tens of picoseconds[19] and recombine. However, this transfer is restricted due to Pauli blocking at high excitation intensities, which results in an increase in the PL height of peak *A* at high excitation intensity. At 77 K [see inset of Fig. 3(a)], however, while the energy levels of QD-*A* and QD-*B* resonate with each other, the resonant near-field interaction becomes weak due to thermal broadening of their energy levels.[20] Therefore, the excitons in each QD relax along with each relaxation path independently.

We analyzed the PL intensity dependence on the excitation intensities to estimate the energy transfer time using a rate equation model. For example, the equation of the $n=2$ state of QD-*B* is given by

$$\frac{dN_2^{\text{QD-}B}}{dt} = -\frac{N_2^{\text{QD-}B}}{\tau} + \frac{N_0^{\text{QD-}B}}{\tau_{\text{rel}}}\frac{D_2 - N_2^{\text{QD-}B}}{D_2} - \frac{N_2^{\text{QD-}B}}{\tau_{\text{rel}}}\frac{D_1 - N_1^{\text{QD-}B}}{D_1} + \frac{N_1^{\text{QD-}A}}{\tau_{\text{et}}}\frac{D_2 - N_2^{\text{QD-}B}}{D_2} - \frac{N_2^{\text{QD-}B}}{\tau_{\text{et}}}\frac{D_1 - N_1^{\text{QD-}A}}{D_1},$$

where $N_i^{\text{QD-}J} = N_i^{\text{QD-}J}(t)$ $(i=1,2)$ $(J=A,B)$ represents the number of carriers in the *i*th energy levels of QD-*J*, and τ_r, τ_{rel}, and τ_{et} are the radiative recombination, the relaxation, and the energy transfer times, respectively. The fitting parameter, τ_{et}, includes the effect of ONF interaction as $U=\hbar/2\tau_{\text{et}}^{-1}$. $N_0^{\text{QD-}J}$ represents the initial state after excitation. In this analysis, we assumed that only the lowest two states of QD-*A* and QD-*B* contribute substantially to the luminescence because we did not observe PL of the $n \geqslant 3$ state under our excitation conditions. Thus, the energy levels of the $n \geqslant 3$ state of QDs and of the GaAs layer after excitation are written as one initial state. The degeneracy of the *i*th level of the QDs, D_i, is $2i$. The term $[D_i - N_i^{\text{QD-}J}]/D_i = f(t)$ represents a filling factor considering that the energy relaxation of carriers or energy transfer can occur only to the unoccupied state of QD-*J* (Pauli blocking). Our model includes the nonradiative path below the $n=1$ state of each QD. As an initial condition, we assumed that twice the number of carriers are captured in QD-*A* as compared to QD-*B*. We used the values of $\tau_r = 1500$ ps and $\tau_{\text{rel}} = 40$ ps.[19] The calculation results are shown in Figs. 3(a) and 3(b) in which the dotted and broken lines represent the integrated PL intensity of the $n=1$ state of QD-*B*, and the total PL intensity of the $n=1$ state of QD-*A* and $n=2$ state of QD-*B*, respectively. These curves agree with the trends of the observed results. From our relatively crude analysis, we estimated the energy transfer times to be 75 ps at 15 K and 4 ns at 77 K. The evaluated τ_{et} at 15 K was much less than τ_r of ~1.5 ns, which was consistent with the experimental observation that PL-peak *A* disappears at low excitation intensity.

A plausible explanation for the strong restriction of energy transfer at 77 K compared to that at 15 K is the broadening of linewidth of the QDs with increases in temperature because the energy transfer rate between QD-*A* and QD-*B* is proportional to the energy overlap of their resonant energy states.[20] We evaluated the increase in τ_{et} with temperature increase from the spectral overlap of single QD-*A* and single QD-*B*. We used a value of 300 μeV as the homogeneous linewidth of a SQD at 15 K.[21] The typical linewidth of QDs broadens to more than 1 meV at 77 K. We estimated that τ_{et} at 77 K increases to 300 ps, a value much smaller than our fitting result. A further possible explanation for the increasing τ_{et} is the breaking of the resonant state by differences in energy shift of QD-*A* and QD-*B* with increasing temperature due to their size dispersion.[22] Further studies are required to clarify the mechanism. The observed fast energy transfer between QDs indicated that III-V compound semiconductor QDs are appropriate for ONF switches.

In summary, we directly observed fast excitation energy transfer between DQDs. By analysis based on the resonant energy transfer via an ONF, the energy transfer time was estimated to be 75 ps at 15 K. Our experimental results indicated that the III-V compound semiconductor QD is an appropriate material for the nanophotonic switch.

This paper is part of the Innovative Nanophotonics Components Development project, under contract with the new Energy and Industrial Technology Development Organization (NEDO) through OITDA.

[1] R. Heitz, I. Mukhametzhanov, P. Chen, and A. Madhukar, Phys. Rev. B **58**, R10151 (1998).
[2] A. Tackeuchi, T. Kuroda, K. Mase, Y. Nakata, and N. Yokoyama, Phys. Rev. B **62**, 1568 (2000).
[3] Yu. I. Mazur, Zh. M. Wang, G. G. Tarasov, M. Xiao, G. J. Salamo, J. W. Tomm, V. Talalaev, and H. Kissel, Appl. Phys. Lett. **86**, 063102 (2005).
[4] Yu. I. Mazur, X. Wang, Z. M. Wang, G. J. Salamo, M. Xiao, and H. Kissel, Appl. Phys. Lett. **81**, 2469 (2002).
[5] C. R. Kagan, C. B. Murray, M. Nirmal, and M. G. Bawendi, Phys. Rev. Lett. **76**, 1517 (1996).
[6] K. Mukai, S. Abe, and H. Sumi, J. Phys. Chem. B **103**, 6096 (1999).
[7] M. Ohtsu and H. Hori, *Near-Field Nano-Optics: From Basic Principles to Nano-Fabrication and Nano-Photonics* (Kluwer, Dordrecht, 1999).
[8] M. Ohtsu, K. Kobayashi, T. Kawazoc, S. Sangu, and T. Yatsui, IEEE J. Sel. Top. Quantum Electron. **8**, 839 (2002).
[9] T. Kawazoe, K. Kobayashi, J. Lim, Y. Narita, and M. Ohtsu, Phys. Rev. Lett. **88**, 067404 (2002).
[10] S. Sangu, K. Kobayashi, A. Shojiguchi, and M. Ohtsu, Phys. Rev. B **69**, 115334 (2004).
[11] T. Kawazoe, K. Kobayashi, S. Sangu, and M. Ohtsu, Appl. Phys. Lett. **82**, 2957 (2003).
[12] T. Kawazoe, K. Kobayashi, K. Akahane, M. Naruse, N. Yamamoto, and M. Ohtsu, Appl. Phys. B: Lasers Opt. **84**, 243 (2006).
[13] T. Yatsui, S. Sangu, T. Kawazoe, M. Ohtsu, S. J. An, J. Yoo, and G.-C. Yi, Appl. Phys. Lett. **90**, 223110 (2007).
[14] W. Wu, J. R. Tucker, G. S. Solomon, and J. S. Harris, Jr., Appl. Phys. Lett. **71**, 1083 (1997).
[15] Z. Y. Xu, Z. D. Lu, X. P. Yang, Z. L. Yuan, B. Z. Zheng, J. Z. Xu, W. K. Ge, Y. Wang, J. Wang, and L. L. Chang, Phys. Rev. B **54**, 11528 (1996).
[16] A. Polimeni, A. Patanè, M. Henini, L. Eaves, and P. C. Main, Phys. Rev. B **59**, 5064 (1999).
[17] M. Gurioli, A. Vinattieri, M. Zamfirescu, M. Colocci, S. Sanguinetti, and R. Nötzel, Phys. Rev. B **73**, 085302 (2006).
[18] Y. P. Varshni, Physica (Amsterdam) **34**, 149 (1947).
[19] S. Malik, E. C. Le Ru, D. Childs, and R. Murray, Phys. Rev. B **63**, 155313 (2001).
[20] Generally, excitation energy transfer between QD-*A* and QD-*B* occurs when the overlap integrals of $\int \rho_A(E)$, $\rho_B(E)dE$ are not zero, where the linewidth of QD-*J* is given by $\rho_J(E) = \hbar\rho^{-1}/[(E-E_J)^2 + (\hbar/2\tau^{-1})^2]$ with lifetime τ.
[21] I. Favero, G. Cassabois, R. Ferreira, D. Darson, C. Voisin, J. Tignon, C. Delalande, G. Bastard, Ph. Roussignol, and J. M. Gerard, Phys. Rev. B **68**, 233301 (2003).
[22] H. Kissel, U. Müller, C. Walther, W. T. Masselink, Yu. I. Mazur, G. G. Tarasov, and M. P. Lisitsa, Phys. Rev. B **62**, 7213 (2000).