

InAs/GaAs Multi-Coupled Quantum Dots Structure Enabling High-Intensity, Near-1.3- μ m Emission due to Cascade Carrier Tunneling

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We have proposed a noble quantum dots system called multi-coupled quantum dots which enables the lateral carrier transfer among quantum dots. In this system, since the photoexcited carriers tunnel into the relatively larger quantum dots, photoluminescence becomes narrower and stronger than in conventional quantum dots. InAs/GaAs self-organized multi-coupled quantum dots has shown strong photoluminescence near 1.3 μ m at room temperature. The cascade-like carrier tunneling among quantum dots has been observed using time resolved photoluminescence. The observed strong dependence of the photoluminescence decay time on the energy has been modeled in good accordance with a simple rate equation assuming a tunneling time of 1300 ps.

Quantum dots or wires built with III-V materials have been proposed for the use in many important electronic and photonic applications.¹ Recent progress in the fabrication of high-quality quantum dots has created opportunities for the detailed experimental studies of zero-dimensional quantum structures.²⁻⁷ However, there is still the problem of size fluctuation during fabrication. This size fluctuation distributes the state density over a wide energy range, resulting in spectrally broad luminescence. This broadening spoils the sharp state density of a quantum dot, which is expected to improve laser performance by using them as the active layer of a semiconductor laser.¹ Also, some theoretical studies^{8,9} have proposed that the photoluminescence (PL) efficiency of quantum dots would be intrinsically poor, due to the severe restriction on the energy and momentum relaxation of carriers; known as the phonon relaxation bottleneck. This effect limits the potential application of the quantum dot structure, especially for semiconductor lasers which require efficient flow and photoemission of the injected carriers. To solve these problems, we have proposed a multi-coupled quantum dots (MCQD) which enhances the PL intensity and reduces the spectral width by utilizing carrier-tunneling between quantum dots. In this paper we report fabrication and characterization of InAs/GaAs MCQD enabling high-intensity near-1.3- μ m emission. We also describe the first observation of cascade-like carrier tunneling among multi-coupled quantum dots.

In the multi-coupled quantum dots, since the dots couple with each other laterally, the wave function of carriers in a quantum dot penetrates into adjacent dots. Photoexcited carriers tunnel and relax into the larger and energetically-lower quantum dots. When the carriers finally reach the local energetic minimum, they radiate there. This process reduces the spectral width of luminescence smaller than the original distribution of the state density and enhances the luminescence intensity. Also, the spread of wave function into the adjacent dots is expected to relieve the phonon relaxation bottleneck by increasing the number of quantum levels relating to carrier relaxation. The InAs/GaAs quantum dots are formed in situ, utilizing the islanding effect of InAs.^{2,3} The self-organization of InAs deposits occurs during strained growth on a flat GaAs buffer layer. InAs, which has a 7 % lattice mismatch to GaAs, is known to deposit first as

a fully-strained two-dimensional layer and then to gradually relax to quantum dots. The InAs layers were grown at a rate of 900 angstroms per hour to a coverage of 2.5 monolayers (ML) at 520 °C under an As pressure of 6×10^{-6} Torr by molecular beam epitaxy on a GaAs buffer. A nominal indium composition of 100 % was used when growing quantum dots. However, due to the complicated growth dynamics of the strained layer, the amount of strain and In content cannot be evaluated directly. For optical measurements, the InAs dots were immediately covered with a GaAs cap layer. For a reference, a conventional quantum dot sample was also grown with InAs deposit of 1.8 ML.

An atomic force microscope image after the deposition of InAs of 2.5 ML indicates that the MCQD has a mean base diameters of 260 Å with a standard deviation of 11 %.¹⁰ The dot density is $8.7 \times 10^{10} \text{ cm}^{-2}$. The quantum dot is conical and the average height is 50 Å. The width of the half-height is 60-70 % that of the base. The average distance to the nearest neighbor in the MCQD structure is 260 Å from center to center. This short distance means that the base of a quantum dot in MCQD makes contact with one adjacent quantum dot's base. Thus, the carriers in quantum dots of MCQD structure seem to have a sufficient tunneling probability to transfer to the adjacent quantum dots. On the other hand, in the conventional quantum dots with InAs deposit of 1.8 ML, the density of dots is $5.4 \times 10^{10} \text{ cm}^{-2}$ and the average distance between quantum dots is 430 Å where the tunneling probability is small.

The tunneling affects the line shape of the CW photoluminescence (PL). Figure 2 shows the CW PL spectra and the PL decay time of MCQD and the conventional quantum dots with InAs deposit of 1.8 ML. The PL decay time was measured by a streak camera with a time resolution of 10 ps. The conventional low-density quantum dots have a single and symmetrical peak. This symmetry can be ascribed to the distribution of the state density originated from the size fluctuation. On the other hand, the PL of the MCQD structure with InAs deposit of 2.5 ML has two peaks as shown in Fig. 2 (b). In MCQD, since a wavefunction of carriers in a quantum dot penetrates into the adjacent dots, photoexcited carriers tunnel and relax into the quantum levels in the larger and energetically-lower quantum dots. Then the carriers finally reach the energetic local minimum and radiate.

This process is expected to reduce the spectral width of PL below that of the original distribution of the state density, and also to enhance the peak PL intensity. The sharp peak at 1.07 eV is ascribed to the luminescence from these local minima, although the high energy peak at 1.14 eV is attributed to the distribution of the state density. Clearly, the intensity of the low-energy peak is improved twice that of the high-energy peak, and the width of the low energy peak becomes narrower than a half of the high-energy peak. At room temperature, the PL wavelength shifts to 1.24 μm . This near-1.3 μm wavelength is very useful for optical-fiber based communication systems.

The cascade-like carrier tunneling between quantum dots is observed in the PL decay time for the first time. The dependence of the PL decay time on the energy contrasts in Fig. 2(a) with Fig. 2(b). Note that in the conventional structure the decay times are around 1250 ps and basically independent of energy. This means that the carrier lifetime in each quantum dots is not sensitive to 11 % size fluctuation. However, in the MCQD structure the decay time strongly depends on the energy, as shown in Fig. 2(b). The decay time is faster for the higher energy levels above 1.1 eV. The energy dependence can be understood systematically by assuming a cascade-like tunneling model. Since the carriers in the higher states have a larger number of lower energy states to be relaxed into, the decay time is expected to be faster for the higher energy. The carriers in the lower energy levels have a longer decay time due to the increase of the carriers relaxed from the higher levels and also due to the decrease of the lower energy levels to be relaxed into. This tunneling and energy relaxation from each energy level to the other occur succeeding like cascade. We assume that the carrier relaxation rate from an energy level to any other levels is proportional to the number of the vacant lower energy levels. Also, we assume for simplicity that the excitation is very weak where carrier occupation in each states is regarded to be very small. In this situation a rate equation for a quantum level, E_j , is written as follows:

$$dn_j / dt = -n_j / \tau_r - \sum_{i>j} n_i D_j / \tau_t + \sum_{i<j} n_i D_i / \tau_t. \quad (1)$$

Here, τ_r is recombination life time, τ_t is tunneling time, and D_j is density of the E_j state. For the initial condition, the carriers are assumed to be distributed over the energy states proportional to the gaussian density state with full width at half maximum of 80 meV. Figure 3 shows the fitting result for $\tau_r = 1200$ ps and $\tau_t = 1300$ ps. The experimental decay time is replotted assuming that the center of the state density is at 1.14 eV. Note that the experimental dependence is explained quite well by this simple model for energy levels higher than -50 meV. This good agreement shows that the PL decay time is governed by tunneling time in addition to recombination lifetime. This is a proof that the carriers actually tunnel laterally and relax into the lower levels succeeding. In the PL experiment, since the decay time is governed by carriers with a shorter lifetime, the observed tunneling time seems to be that of electrons whose effective mass is smaller than that of holes. The ratio of the recombination lifetime over the tunneling time revealed that tunneling occurs 0.9 times on average during a carrier lifetime. This repetition rate of tunneling will be

improved by reducing the tunneling time. In energy levels lower than -50 meV, the theoretical fitting does not agree with the experimental data. The calculated decay time of 1700 ps for the lower energy levels is longer than that of the experimental results. In these lower energies, the experimental decay time becomes shorter for lower energy levels. Since the excitation power is assumed to be very weak in calculation, the model does not agree with the highly populated levels under 1.1 eV where the PL becomes stronger as shown in Fig. 2 (b). In this region where the carrier density is relatively large in comparison with the state density, stimulated emission with short lifetime might partially occur.

In application, the wavelength of the luminescence is interesting because the wavelength is 1.24 μm at room temperature. This near-1.3- μm wavelength can be very useful practical optical-fiber-based communication systems. Furthermore, this luminescence is obtained on a GaAs substrate which has wider energy band gap than the InP substrate. Conventional 1.3 μm optical devices are formed on InP substrates which are more leaky than GaAs substrates due to their lower band gap energy. In order to evaluate the applicability of this material to actual devices, we have compared its intensity with that of the strained InGaAs/GaAs QWs whose luminescence wavelength is around 0.98 μm . The InGaAs/GaAs quantum well is known to show an extremely low threshold current density of as low as 450 A/cm² in the application to surface-emitting diode lasers.¹¹ Both the photoluminescence intensities at room temperature in In_{0.2}Ga_{0.8}As/GaAs QWs and InAs/GaAs MCQD are shown in Fig. 4 after the normalization by the best datum for the quantum wells. The intensity of the MCQD is 4 % of the best datum for InGaAs/GaAs QWs grown at 540 °C. However, note that this is higher than the lowest datum for quantum wells; it is 150 % of the lowest one grown at 570 °C. The intensity of InGaAs/GaAs QWs strongly depends on the growth temperature. This result shows that the MCQD has a luminescence intensity comparable with that of quantum wells. Also, the luminescence intensity will probably be improved by further optimization of the growth conditions. If the intensity is transformed to the value per effective volume, the datum for the quantum dot is increased to 40 % (1400 %) of the best (lowest) datum for quantum wells, which indicates the high quality of these dot samples. The InAs/GaAs MCQD thus shows high-intensity photoluminescence at 1.24 μm at room temperature, comparable to the well-known highly efficient In_{0.2}Ga_{0.8}As/GaAs QWs.

In summary, we have proposed a noble quantum dots system called multi-coupled quantum dots which enables the lateral carrier transfer among quantum dots. The MCQD reduce the photoluminescence width, with the help of the tunneling, narrower than the distribution of the density of states which originated from the size fluctuation of quantum dots. InAs/GaAs self-organized MCQD show strong photoluminescence near 1.3 μm at room temperature which is very useful wavelength in practical application. The cascade-like carrier tunneling among quantum dots has been observed for the first time using time resolved photoluminescence. The observed strong dependence of the photoluminescence decay time on the

energy has been modeled in good accordance with simple rate equation assuming a tunneling time of 1300 ps.

We thank Drs. Teruo Sakurai and Hajime Ishikawa for their encouragement. We also acknowledge Dr. Motomu Takatsu for useful discussion.

References

- 1) Y. Arakawa and H. Sakaki: Appl. Phys. Lett. 40 (1982) 939.
- 2) J. Y. Marzin and J. M. Gerard: Superlattices and Microstruct. 5 (1989) 51.
- 3) R. Nötzel, J. Temmyo and T. Tamamura: Nature 369 (1994) 131.
- 4) J. M. Moison, F. Houzay, F. Barthe, L. Leprince, E. Andre and O. Vatel: Appl. Phys. Lett. 64 (1994) 196.
- 5) S. Fafard, D. Leonard, J. L. Merz and P. M. Petroff: Appl. Phys. Lett. 65 (1994) 1388.
- 6) J. Oshinowo, M. Nishioka, S. Ishida and Y. Arakawa: Appl. Phys. Lett. 65 (1994) 1421.
- 7) K. Mukai, N. Ohtsuka, M. Sugawara and S. Yamazaki: Jpn. J. Appl. Phys. 33 (1994) L1710.
- 8) U. Bockelmann and G. Bastard: Phys. Rev. B42 (1990) 8947.
- 9) H. Benisty, C. M. Sotomayortorres and C. Weisbuch: Phys. Rev. B44 (1991) 10945.
- 10) A. Tackeuchi, Y. Nakata, S. Muto, Y. Sugiyama, and N. Yokoyama: Jpn. J. Appl. Phys. 34 (1995) L405.
- 11) H. Shoji, K. Otsubo, M. Matsuda and H. Ishikawa: Electron. Lett. 30 (1994) 409.

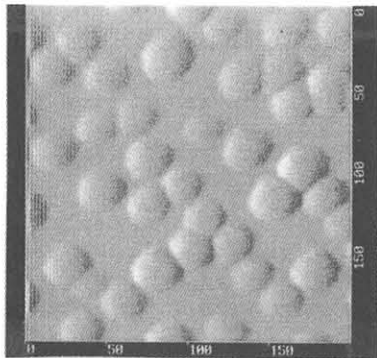


Fig. 1. AFM image of InAs/GaAs MCQD (InAs deposition : 2.5 ML) before overgrowth. The scanned area is 200 nm x 200 nm.

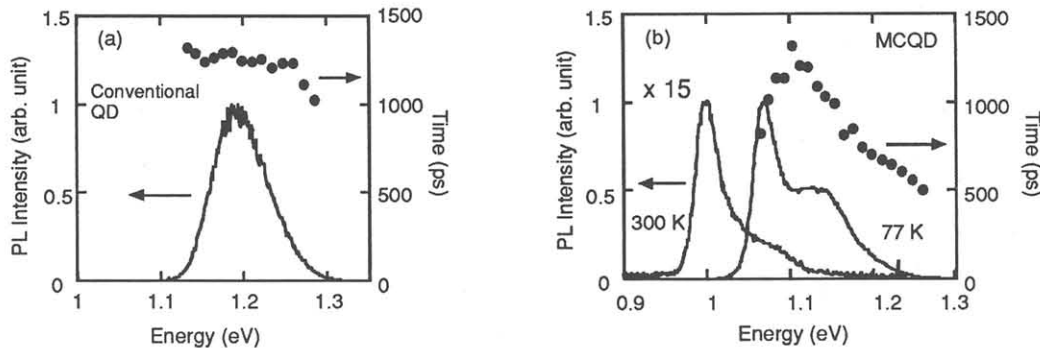


Fig. 2. (a) CW PL spectra of conventional quantum dots (InAs deposition : 1.8 ML) for the excitation power of 0.6 mW at 77 K. The PL decay time for the excitation power of 5 mW at 77 K. (b) CW PL spectra of MCQD for the excitation power of 2.5 mW at 77 K. The PL decay time for the excitation power of 5 mW at 77 K.

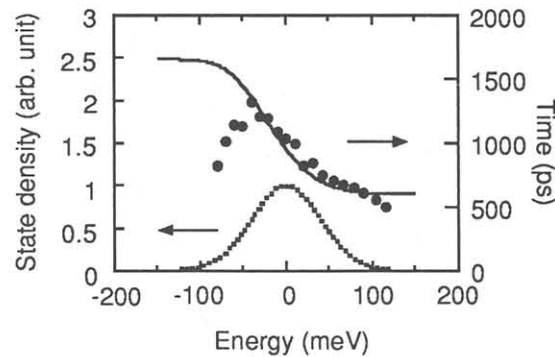


Fig.3. Fitting by rate-equation (solid line) assuming a gaussian shape state density (dotted line) with a full width at half maximum of 80 meV. The experimental PL decay times for the MCQD are replotted by the solid circles.

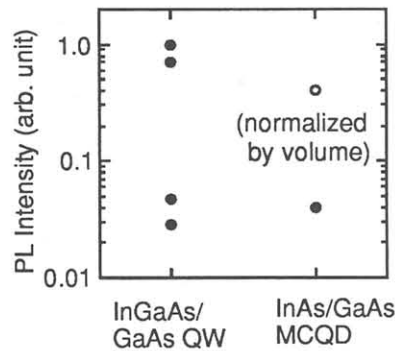


Fig. 4. PL intensity of MCQD and InGaAs/GaAs quantum wells (QW) at room temperature. The intensities are normalized by the best datum for InGaAs/GaAs QW grown at 540 °C. The open circle shows the relative intensity per effective volume of MCQD.