Lasing in GaAs/AlGaAs self-assembled quantum dots

T. Mano, T. Kuroda, M. Yamagiwa, G. Kido, K. Sakoda et al.

Citation: Appl. Phys. Lett. **89**, 183102 (2006); doi: 10.1063/1.2372448
View online: http://dx.doi.org/10.1063/1.2372448
View Table of Contents: http://apl.aip.org/resource/1/APPLAB/v89/i18
Published by the American Institute of Physics.

Lasing in GaAs/AlGaAs self-assembled quantum dots

T. Mano,[a)] T. Kuroda,[b)] M. Yamagiwa, G. Kido, K. Sakoda,[c)] and N. Koguchi
Quantum Dot Research Center, National Institute for Materials Science, 1-2-1 Sengen, Tsukuba, Ibaraki 305-0047, Japan

(Received 28 August 2006; accepted 10 September 2006; published online 30 October 2006)

The authors have demonstrated photopumped laser action of self-assembled ring-shaped GaAs quantum dots (QDs) grown by droplet epitaxy. Morphological control of the QD shape from conelike dots to ringlike ones results in a narrow spectral band of photoluminescence from the QD ensemble, reflecting their small size distribution. Using ring-shaped QDs as an active laser medium, they observed multimodal stimulated emissions from the ground state at temperatures of up to 300 K. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2372448]

The GaAs/AlGaAs heterostructure is a prototype semiconductor system that is used in a variety of novel electronic and photonic devices. One advantage of this system is the fact that GaAs/AlGaAs is a lattice-matched system that allows for ideal heteroepitaxial growth. The drawback is the difficulty in producing self-assembled quantum dots (QDs) using the widely applied strain-induced Stranski-Krastanow growth technique. On the other hand, it has been recognized that the use of QDs as the active medium of lasers has numerous merits that are not available in bulks or quantum wells (QWs).[1,2] For example, high-speed modulation, low-threshold current density, and high-temperature characteristics have been demonstrated.[3,4] Thus, from both practical and fundamental standpoints, it is of immense interest to develop QD laser structures using the GaAs/AlGaAs heterosystem.

We have proposed *droplet epitaxy* which is an alternative self-assembling technique for forming QDs.[5–7] The mechanism of this technique is based on the self-assembly of liquid metal nanoparticles (droplets) of group-III atoms and their crystallization into III-V semiconductor QDs by a subsequent supply of group-V atoms.[6] This technique has been successfully applied to both lattice-matched and lattice-mismatched compound-semiconductor systems.[5–9] In this letter we report on the photopumped laser operation in self-assembled GaAs/AlGaAs QDs. Although laser emission in submicron conical-shaped GaAs microstructures had previously been reported, these are regarded not as QDs but as QWs (or disks).[10] A key for achieving laser action in GaAs QDs is an improvement in their size and shape uniformity. We have recently observed a morphological change of QDs from a conelike shape to a ringlike one in the droplet epitaxy.[11,12] In the case of ring-shaped QDs, we found a great reduction in the photoluminescence (PL) spectral width, reflecting the small size distribution of the QDs. The narrow spectral feature results in high optical gain for stimulated emissions. The laser operation was confirmed at temperatures of up to 300 K.

The sample was grown on a semi-insulating GaAs (100) substrate by a conventional solid-source molecular-beam epitaxy system. The QD-laser structure consists of (1) a 1.3-μm-thick $Al_{0.58}Ga_{0.42}As$ bottom cladding layer, (2) a 0.28-μm-thick $Al_{0.23}Ga_{0.77}As$ active layer with three-stacked ring-shaped GaAs QDs (separated by 20-nm-thick $Al_{0.23}Ga_{0.77}As$),[13] and (3) a 1.3-μm-thick $Al_{0.58}Ga_{0.42}As$ top cladding layer. The ring-shaped QDs in the active layer were formed by droplet epitaxy. On the $Al_{0.23}Ga_{0.77}As$-$c(4\times4)$ surface, nominally 4.75 ML Ga (1.5 ML/s) was supplied without As_4 flux at 200 °C for the droplet formation. During the Ga supply, the first 1–2 ML of Ga was combined with excess As atoms on the $c(4\times4)$ surface, forming a two-dimensional GaAs layer, and the rest of the Ga formed droplets.[6,14] These droplets were then crystallized into ring-shaped GaAs QDs by supplying an As_4 flux (1×10^{-5} Torr beam equivalent pressure) at the same temperature.[11] The QDs were annealed at 350 °C for 10 min under As_4 flux supply without capping to improve the crystal quality. After annealing, the QDs were covered with a 15-nm-thick AlGaAs capping layer at 350 °C and the rest of the AlGaAs layer was grown at 580 °C. Once the entire growth sequence was completed, a rapid thermal annealing process was performed (800 °C for 4 min) in a N_2 atmosphere to improve the optical quality.[12,15]

Figure 1(a) shows an atomic force microscopy image of the surface followed by annealing at 350 °C for 10 min. Well-defined ring-shaped QDs are present with high spatial density: the density of QDs is estimated to be $2\times10^{10}/cm^2$. A cross-sectional profile of a typical QD is shown in Fig. 1(b). Their average top diameter and height are 27 and 2 nm, respectively. The QDs show a size fluctuation of 13% in diameter and 27% in height.

Figure 2 shows PL spectra of the sample at stationary excitation. The PL was detected from the top of the sample. At 5 K, an intense PL peak of carrier recombination in QDs was observed at 741 nm (1.67 eV), together with small signals at 682 nm (1.80 eV) and 688 nm (1.82 eV) arising from a two-dimensional GaAs layer and/or an (Al, Ga)As barrier. The QD PL peak well fits a Gaussian function with 34 meV in full width at half maximum (FWHM). This value is nearly one-third of that of the cone-shaped QDs which we have previously reported,[7,15] suggesting high uniformity in the size and shape of the ring-shaped QDs.

The PL emission was clearly observed up to room temperature (RT) as seen in the lower panel of Fig. 2, which shows a doublet QD spectrum consisting of a major peak at 780 nm (1.59 eV) and a minor one at 752 nm (1.65 eV).

[a)]Author to whom correspondence should be addressed; electronic mail: mano.takaaki@nims.go.jp
[b)]Also at: PRESTO, Japan Science and Technology Agency.
[c)]Also at: Graduate School of Pure and Applied Sciences, University of Tsukuba.

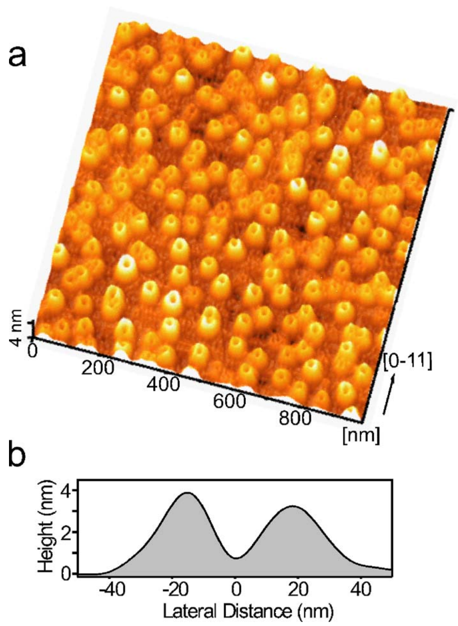

FIG. 1. (Color online) (a) Atomic force microscope image of ring-shaped GaAs QDs. The scan area is $1\times1\ \mu m^2$. (b) Cross-sectional profile of a ring-shaped QD.

Appearance in the higher energy peak should be related to the thermal activation of photocarriers into the excited state of QDs. Accordingly, we can identify the excited state of our QDs being present at ~0.06 eV higher than the ground state. The relevant energy split agrees with a theoretical value based on the effective-mass approximation calculation.[16]

For the laser emission experiments, we cleaved the sample into a rectangular piece, presenting a laser cavity of 2200 μm in longitudinal length. Both of the cleaved facets were uncoated. For excitation, we used a frequency-doubled *Q*-switched neodymnium yttrium aluminum garnet (Nd-YAG) laser, producing pulses of 532 nm in wavelength, 5 ns in duration, and 4 kHz in repetition. The laser beam was focused into a single stripe on the top surface of the sample using a pair of cylindrical lenses. The width of stripe excitation was 20 μm, corresponding to transverse length in the cavity. The emission normal to the cleaved surface was detected by a spectrometer of 50 cm focal length equipped with a charge-coupled device, whose spectral resolution was 0.07 nm.

Figure 3(a) shows emission spectra taken from the cleaved edge of the laser structure for various excitation

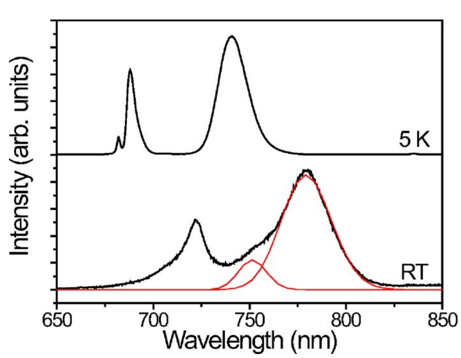

FIG. 2. (Color online) PL spectra of an ensemble of ring-shaped QDs measured at 5 K (upper) and RT (lower). A fit to the RT spectrum using two Gaussian functions is also shown.

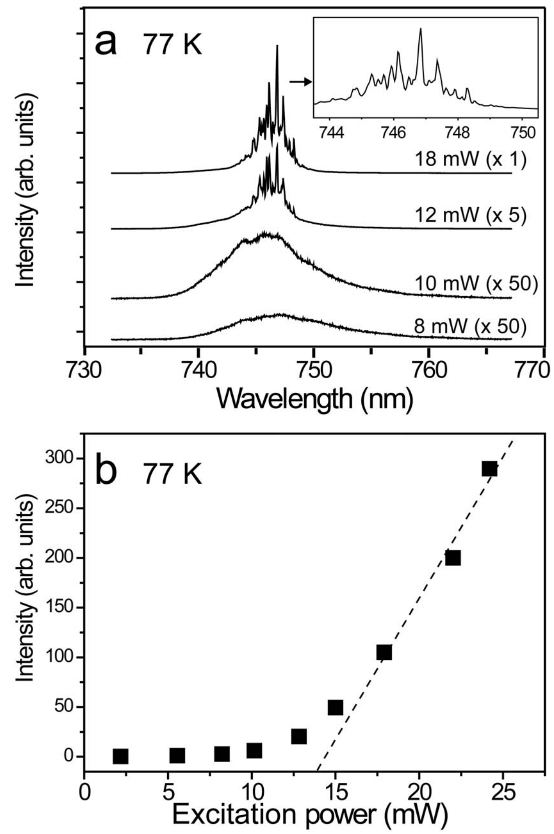

FIG. 3. (a) Emission spectra taken from the cleaved side with various excitation powers (8, 10, 12, and 18 mW) at 77 K. The inset shows a magnified spectrum at 18 mW. (b) Integrated emission intensities at 77 K as a function of excitation power.

powers at 77 K. At low excitation below 8 mW, the spectra show a smooth and broad curve corresponding to spontaneous emissions from the QD ensemble. When the excitation power increases to 12 mW, the spectra change into an ensemble of sharp lines, reflecting multimodal stimulated emissions of QDs. The FWHM of each line is limited by instrumental resolution. The center energy of the line ensemble is 747 nm, which agrees with that of spontaneous emissions. This observation suggests that stimulated emissions originated from the ground state of QDs. Figure 3(b) is a plot of the dependence of emission intensity on excitation power: The intensity was evaluated by the spectral integration of emission signals. We can observe a clear threshold that means the onset of lasing. The inset in Fig. 3(a) shows an expanded view of the multimodal laser spectrum. Emission peaks are quasiperiodically separated by $\Delta\lambda \sim 0.2$ nm. In our device, however, the longitudinal mode separation is expected to be ~0.03 nm, according to a well-known expression, $\Delta\lambda=\lambda^2/(2nL)$, where λ (emission wavelength) =750 nm, n (refractive index)=4, and L (cavity length) =2200 μm. Thus, the line ensemble in the spectrum does not correspond to longitudinal modes, which are unresolved by our equipment. We believe that the observed spectra are related to a distinct group of longitudinal modes exceeding the threshold. Similar behavior has been observed in a variety of QD-based lasers.[17,18]

We investigated temperature dependence of the lasing properties to check the quality of our device. Figure 4(a) shows the laser emission spectra at various temperatures. Note that the spectral resolution in this measurement was

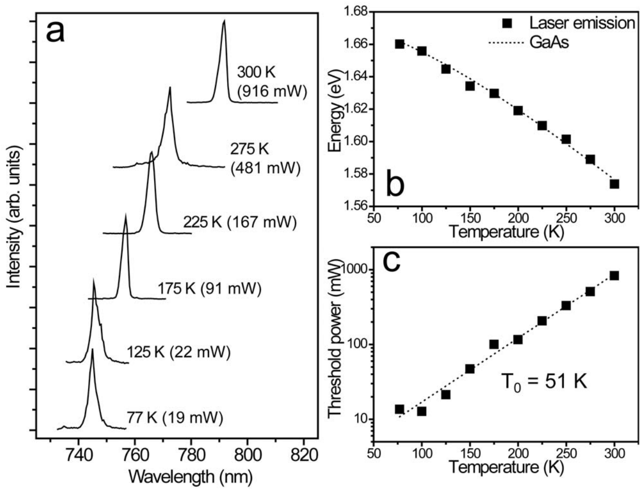

FIG. 4. (a) Lasing spectra at 77, 125, 175, 225, 275, and 300 K. [(b) and (c)] Center energy and threshold power of the lasing as a function of temperature. The dotted line in (b) shows the temperature dependence of the GaAs band gap energy. The dotted line in (c) shows the fit according to the characteristic temperature of 51 K.

5 nm, so that the multimodal structure was smoothed out. The spectra of laser emissions shifted to lower energy with increasing temperature. Figure 4(b) is a plot of the temperature dependence of the center energy of laser emissions, which shows a monotonic decrease with temperature. It follows the GaAs band gap energy, as shown by the dotted line in Fig. 4(b). Thus, we can conclude that the electronic level relavent to the lasing is the ground state of QDs irrespective of temperature up to RT.

Temperature dependence of threshold power is shown in Fig. 4(c). Although the threshold power increased with temperature, laser action was confirmed up to RT, indicating the good quality of our device. Through the fit, we estimated the characteristic temperature (T_0) to be 51 K. This value is not as good as the hypothetical ideal value for QD lasers.[1] We attribute this mainly to the small band offset between the GaAs QDs and $Al_{0.23}Ga_{0.77}As$ barrier, which caused effective thermal carrier escape from the QDs to the barrier layers.[19] We may improve it by using AlGaAs with higher Al composition.

In summary, we demonstrated photopumped lasing from ring-shaped GaAs QDs grown by droplet epitaxy. By making use of ring-shaped QDs, we achieved a great reduction in PL spectral width, reflecting the small size distribution of the QDs. A QD-laser structure with three layers of these QDs exhibited multimode laser emission from the ground state of the QDs with a clear threshold from 77 K to RT. The observed multimode emission peaks were related to the groups of longitudinal modes. The characteristic temperature of the device was 51 K, which was mainly due to the small band offset. With further optimization of growth processes and further studies of particular electronic states of ring-shaped QDs, droplet epitaxy will be a promising method to produce QD-lasers in lattice-matched compound semiconductor systems.

The authors wish to acknowledge T. Noda, A. Ohtake, K. Mitsuishi, and M. Kawabe for their useful discussions. This work was partly supported by a Grant-in-Aid from the Ministry of Education, Culture, Sports, Science and Technology of Japan.

[1] Y. Arakawa and H. Sakaki, Appl. Phys. Lett. **40**, 939 (1982).
[2] M. Asada, Y. Miyamoto, and Y. Suematsu, IEEE J. Quantum Electron. **QE-22**, 1915 (1986).
[3] P. G. Eliseev, H. Li, A. Stintz, G. T. Liu, T. C. Newell, K. J. Malloy, and L. F. Lester, Appl. Phys. Lett. **77**, 262 (2000).
[4] K. Otsubo, N. Hatori, M. Ishida, S. Okumura, T. Akiyama, Y. Nakata, H. Ebe, M. Sugawara, and Y. Arakawa, Jpn. J. Appl. Phys., Part 2 **43**, L1124 (2004).
[5] N. Koguchi, S. Takahashi, and T. Chikyow, J. Cryst. Growth **111**, 688 (1991).
[6] N. Koguchi and K. Ishige, Jpn. J. Appl. Phys., Part 1 **32**, 2052 (1993).
[7] K. Watanabe, N. Koguchi, and Y. Gotoh, Jpn. J. Appl. Phys., Part 2 **39**, L79 (2000).
[8] T. Mano, K. Watanabe, S. Tsukamoto, N. Koguchi, H. Fujioka, M. Oshima, C. D. Lee, J. Y. Leem, H. J. Lee, and S. K. Noh, Appl. Phys. Lett. **76**, 3543 (2000).
[9] Y. Nonogaki, T. Iguchi, Y. Fujiwara, and Y. Takeda, Appl. Surf. Sci. **117**, 665 (1997).
[10] T. Egawa, A. Ogawa, T. Jimbo, and M. Umeno, Jpn. J. Appl. Phys., Part 1 **37**, 1552 (1998).
[11] T. Mano and N. Koguchi, J. Cryst. Growth **278**, 108 (2005).
[12] T. Mano, T. Kuroda, S. Sanguinetti, T. Ochiai, T. Tateno, J. S. Kim, T. Noda, M. Kawabe, K. Sakoda, G. Kido, and N. Koguchi, Nano Lett. **5**, 425 (2005).
[13] H. Shoji, Y. Nakata, K. Mukai, Y. Sugiyama, M. Sugawara, N. Yokoyama, and H. Ishikawa, Appl. Phys. Lett. **71**, 193 (1997).
[14] A. Ohtake, P. Kocan, J. Nakamura, A. Natori, and N. Koguchi, Phys. Rev. Lett. **92**, 236105 (2004).
[15] K. Watanabe, S. Tsukamoto, Y. Gotoh, and N. Koguchi, J. Cryst. Growth **227**, 1073 (2001).
[16] T. Kuroda, T. Mano, T. Ochiai, S. Sanguinetti, K. Sakoda, G. Kido, and N. Koguchi, Phys. Rev. B **72**, 205301 (2005).
[17] L. Harris, D. J. Mowbray, M. S. Skolnick, M. Hopkinson, and G. Hill, Appl. Phys. Lett. **73**, 969 (1998).
[18] A. Patane, A. Polimeni, L. Eaves, M. Henini, P. C. Main, P. M. Smowton, E. J. Johnston, P. J. Herrmann, G. M. Lewis, and G. Hill, J. Appl. Phys. **87**, 1943 (2000).
[19] A. R. Goodwin, J. R. Peters, M. Pion, G. H. B. Thompson, and J. E. A. Whiteaway, J. Appl. Phys. **46**, 3126 (1975).