AIP Applied Physics Letters

High-Q resonance modes observed in a metallic nanocavity

R. Takemoto, N. Ishihara, H. Kurosawa, N. A. Jahan, T. Asano, X. Liu, H. Nakajima, H. Kumano, and I. Suemune

Citation: Applied Physics Letters **103**, 191104 (2013); doi: 10.1063/1.4828351
View online: http://dx.doi.org/10.1063/1.4828351
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/103/19?ver=pdfcov
Published by the AIP Publishing

High-Q resonance modes observed in a metallic nanocavity

R. Takemoto,[a] N. Ishihara, H. Kurosawa, N. A. Jahan, T. Asano, X. Liu, H. Nakajima, H. Kumano, and I. Suemune[b]
Research Institute for Electronic Science, Hokkaido University, Sapporo 001-0021, Japan

(Received 12 September 2013; accepted 16 October 2013; published online 4 November 2013)

Metallic nanocavities have been actively studied for realizing nanolasers with low threshold. Presence of resonance modes with high cavity Q values is the indication of low internal loss that leads to low threshold lasing. However, cavity Q values observed in metallic nanocavities below lasing threshold remain low at present on the order of 100 to 500. We study the possibility to realize higher resonance Q values with a metallic nanocavity. For probing purpose of cavity modes we propose to employ broad mid-gap-state optical emission of n-type GaAs. With this method we report the observation of a resonance mode with the high Q value of 3800 at room temperature with the metallic nanocavity. The cavity mode is identified as a whispering-gallery mode with finite-element-method simulation. © *2013 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4828351]

In 1960, the first laser operation was demonstrated by Theodore Maiman with a ruby laser.[1] Then through the technology evolution such as laser designs, gain materials, and fabrication processes, semiconductor lasers have been developed to be applied to various fields. Familiar application fields are compact-disc audio players, bar-code readers, optical-fiber communication, and so on. Laser gain medium initiated from p-n homo-junctions, then evolved to double heterostructures, quantum wells, and quantum dots (QDs). Laser structures also evolved from Fabry-Perot lasers to distributed feedback lasers, vertical-cavity surface emitting lasers, and recently to various microcavity lasers.

Following these evolutions, downsizing to nanolasers for lower threshold is one of the recent main topics. This is because nanolasers are expected to be applied for highly integrated photonic circuits,[2] low-energy consumption applications,[2] compact pickups for data storage,[3] biological and chemical sensing,[4] and imaging.[5] Therefore various kinds of nanolasers, especially metal-coated nanolasers attract high attention in recent years.[6–16] Up to now lasing with low threshold,[6,8,13,16] lasing at room temperature,[7,8,10,12,15] and continuous-wave (CW) laser operation[7,12] have been demonstrated. Almost all of them are designed with small sizes of about 1 μm or less three-dimensionally, and therefore the metal-coated nanolasers have extremely small volumes.

These nanolasers are also regarded as metal-coated nanocavities, and the optical quality can be evaluated with their (cold) cavity Q values. As is well known, the Q value is defined as the stored energy in a cavity multiplied by the resonant angular frequency and divided by the internal loss in the cavity. The cavity Q values in these nanolaser structures have been evaluated below the lasing threshold. The measured Q values were in the range of 100 to 500 and remained relatively low.[6,8,10,13,15,17] From the formula of the Q value, higher cavity Q indicates the lower internal loss in the cavity and will lead to the lower lasing threshold. In this regard, realization of higher cavity Q values is a prerequisite for realizing superior laser performance. With simulations, higher Q values have been predicted. Depending on the cavity structures and selection of metals, cavity Q values around 3000 have been estimated. However, these high cavity Q values for metal-coated nanocavities still remain only with simulations at present and have not yet been realized.

In this paper, we experimentally study the possibility of realizing a metal-coated nanocavity with the high Q value. For this purpose, we propose to employ the broad-band room temperature emission of n-type GaAs. We report the observation of the high cavity Q value of 3800 with the metallic nanocavities at room temperature. We also work on finite-element method (FEM) simulations to identify the high-Q cavity mode.

In relation to metallic nanocavities, we have developed single-photon sources with semiconductor QDs embedded in metal. An InAs QD/GaAs heterostructure was etched into a pillar structure and was covered with silver (Ag), and then the GaAs substrate was removed with cleavage.[18] Employing this structure, we worked on the autocorrelation measurements on photons emitted from a single QD in the GaAs pillar that was embedded in Ag. We observed clear anti-bunching dip at zero delay of the two optical detection paths and therefore demonstrated single-photon emission.[18,19] With this metallic nanostructure, we also demonstrated the high single-photon extraction efficiency of ~18%. It is well known that photon extraction efficiency from a planar semiconductor surface is limited to less than 2% due to the total internal reflection at the air/semiconductor interface, and therefore this is a drastic improvement. We study the possibility of this metallic nanostructure whether it can be a nanocavity.

For the purpose of studying cavity resonance modes, we propose to employ mid-gap-state optical emission of n-type GaAs. Figure 1(a) is the comparison of photoluminescence (PL) spectrum measured on three-kinds of (001) GaAs substrates at room temperature. A He-Ne laser at the wavelength of 632.8 nm was used for the excitation. The PL spectrum observed from a semi-insulating (SI) GaAs (upper) is almost noise level. The PL spectrum observed from a p-type GaAs (middle) is dominated by the near-band-gap emission. The

[a]Electronic mail: r.takemoto@es.hokudai.ac.jp
[b]Electronic mail: isuemune@es.hokudai.ac.jp

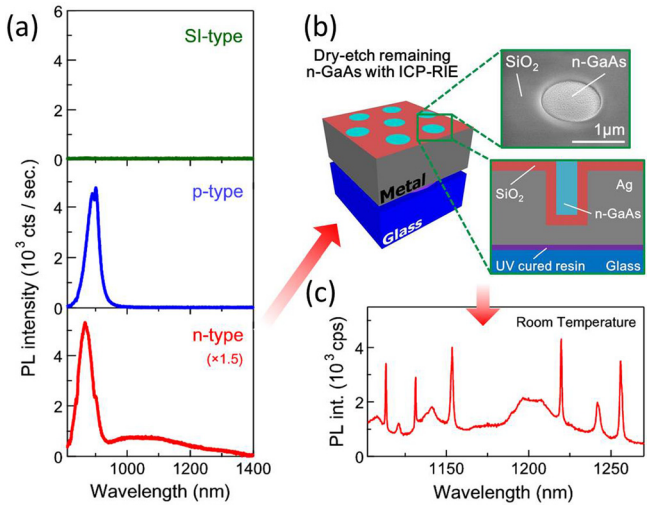

FIG. 1. (a) Comparison of PL spectrum measured on SI-type (upper), p-type (middle), and n-type (lower) GaAs substrates at room temperature. (b) Schematic of n-GaAs pillar embedded in Ag (left), SEM image of n-GaAs pillar surface embedded in Ag (right upper), and the cross-sectional view of the Ag-embedded structure (right lower). (c) PL spectrum measured at room temperature on the n-type GaAs pillar embedded in Ag.

hole concentration is $\sim 1 \times 10^{19}\,\mathrm{cm}^{-3}$ with zinc doping. On the other hand, the PL spectrum measured on an n-type GaAs (lower) exhibits very broad luminescence in the mid-gap region up to the wavelength of 1400 nm. We propose to employ this broad-band room temperature optical emission of n-GaAs to probe cavity modes. The origin of this distinct mid-gap emission is not clear, but the electron concentration is the high level of $\sim 2 \times 10^{18}\,\mathrm{cm}^{-3}$ with silicon (Si) doping. Although Si is prominently incorporated into the Ga site and forms donor levels in GaAs, it is also incorporated into the As site depending on the growth condition and can form acceptor levels.[20] This amphoteric nature of Si originating from the incorporation of the IV-column atom into the III-V semiconductor can result in the possibility to form donor-acceptor pairs in GaAs and related complexes that give the mid-gap optical emission.

We fabricated n-type GaAs pillars embedded in Ag following the same fabrication process as in Ref. 18 and examined its possibility to function as a nanocavity. Employing the n-GaAs mid-gap broad luminescence as the probe, we really observed a cavity resonance PL peak. However, the cavity Q value estimated from the full-width at half maximum (FWHM) of the PL peak was only 80 and remained low (not shown). Our detailed secondary-electron microscope (SEM) observations of the sample revealed that the GaAs pillar surfaces showed some surface structures induced during the cleavage process. This will scatter photons confined inside the cavity and will reduce the cavity Q value. Therefore in order to achieve higher Q values, we tried to improve the substrate removal process. First, we covered the n-type GaAs pillars with a 200-nm-thick SiO_2 layer and subsequent metallization with Ag. Then we changed the GaAs substrate removal process from cleavage to mechanical polishing and the subsequent etching with inductively coupled plasma (ICP)-reactive ion etching (RIE). Figure 1(b) shows the schematic structure and the SEM image of the completed sample. The center circle in the SEM image is the n-type GaAs pillar surface and the other surrounding area is the SiO_2 one. As shown in Fig. 1(b), we could prepare the smooth planar sample surface with this improved fabrication process employing ICP-RIE. We point out two reasons to cover the semiconductor pillars with the SiO_2 layer. One reason is to protect the Ag surface during ICP-RIE of GaAs with chlorine gas (Cl_2), because the etching almost stops at the GaAs/SiO_2 interface. Another reason is to prevent the light penetration from the GaAs pillar into the metal interface to minimize optical absorption loss.

We studied the optical properties of the prepared sample at room temperature. As is shown in Fig. 1(a), the mid-gap emission from an n-type GaAs bulk substrate is broad. When an n-GaAs pillar is embedded in Ag, sharp PL peaks were observed as shown in Fig. 1(c). This pillar has the sidewall taper angle of 6.9° with the surface and bottom diameters of 1170 nm and 1000 nm, respectively, and the height of 700 nm. For this measurement, the He-Ne laser light was focused to a target pillar employing an object lens with the numerical aperture of 0.4. The luminescence from the n-GaAs pillar was collected with the same objective lens and was dispersed with a monochromator. We observed similar sharp peaks with many other pillars with different diameters and different heights. Since no exciton emission is possible below energy gap, these sharp peaks are identified as the luminescence enhanced by cavity modes at room temperature.

We calculated the cavity Q values from the FWHM of the PL peaks and plotted them with the circles with respect to the n-GaAs pillar diameter as shown in Fig. 2. Several cavity modes were observed for a given pillar diameter and the data measured on several pillars are plotted for each pillar diameter in Fig. 2. The red solid line is a guide to the eyes, showing the general tendency of the higher Q value for the larger pillar diameter. We worked on the FEM simulation of the electromagnetic fields in the metal-embedded structure using the eigenmode solver of commercial software COMSOL. In our simulation the refractive indices of n-GaAs and SiO_2 were set to 3.5 and 1.46, respectively. The permittivity of Ag was taken from the literature.[21] We could find two kinds of cavity modes. One is the whispering gallery (WG) mode circulating along the GaAs pillar cross-sectional circumference shown in Fig. 3(a) (with the

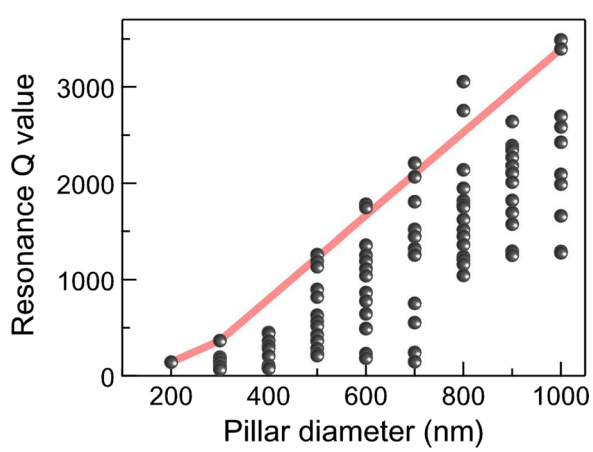

FIG. 2. Plot of measured cavity Q values of the resonance modes for each pillar diameter.

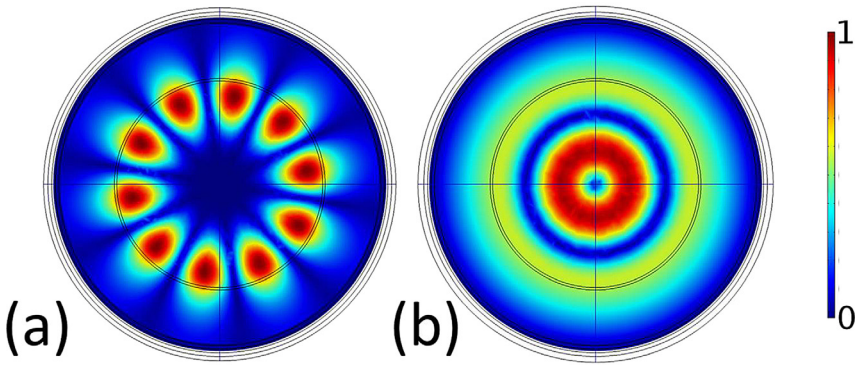

FIG. 3. Electric-field intensity distributions of two resonant modes calculated with FEM simulations. (a) WG mode with the mode number of 5 and (b) TE_{01} mode. 700-nm of n-GaAs pillar diameter is assumed in this calculation and the thin solid circle in each figure shows the boundary of n-GaAs and SiO_2 at the radius of 350 nm. The outer boundary of 200-nm-thick SiO_2 and Ag is assumed at the radius of 550 nm.

mode number of 5). The other is the transverse electric (TE) mode (TE_{01} mode in Fig. 3(b)). The simulation parameters are 700 nm of the GaAs pillar diameter and 700 nm of the pillar height. The WG mode is found at the resonance wavelength of 1020 nm with the cavity Q value of ~500. The TE_{01} mode is found at the resonance wavelength of 1360 nm with the cavity Q value of ~1300. The major part of the modes for given pillar diameters observed in Fig. 2 is mostly the higher-order modes of these two kinds of cavity modes. The observed general tendency of the higher Q values for the larger pillar diameter is difficult to predict directly with the simulations. However for the larger pillar diameter, WG modes with the larger mode numbers are allowed and their cavity Q values generally increase for the larger mode numbers (not shown). This simulation result is consistent with the observation in Fig. 2.

We examined the optical properties of the sharp peaks in more detail by measuring pillars on several samples and focused to a peak at the wavelength of 999.6 nm, which is found with a pillar that has the n-type GaAs pillar height of 800 nm and the pillar surface and bottom diameters of 1200 nm and 1100 nm, respectively, with the sidewall inclination angle of 4.8°. We measured the excitation power dependence of the PL FWHM, since PL peaks are generally broadened for the higher excitation power. The PL FWHM was 0.26 nm and remained almost constant against the increase of the excitation power as shown in Fig. 4. This result also confirms that the observed sharp peak is the cavity mode. The observed sharp peak spectrum was well fitted with the Lorentzian function as shown in the inset of Fig. 4. From the FWHM, the resonance mode is found to have the high cavity Q value of 3800. This is an extremely high Q value observed among metallic nanocavities.

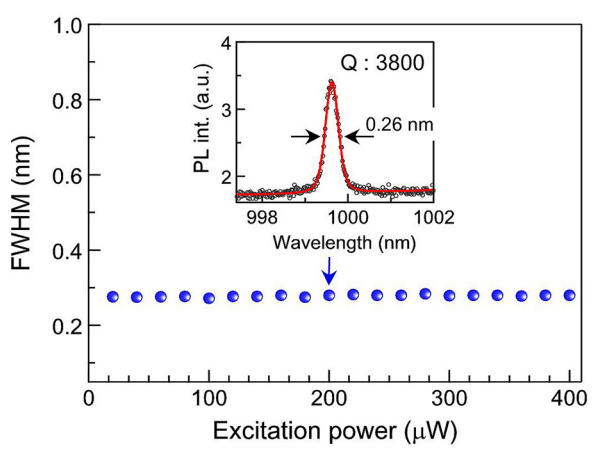

FIG. 4. Excitation power dependence of FWHM of the resonance mode spectrum at the wavelength of 999.6 nm, and the Lorentzian function fit of the PL spectrum (inset).

To identify the origin of this high-Q resonance mode, we worked on the FEM simulation. The thickness of the SiO_2 layer shown schematically in Fig. 1(b) is 200 nm, the n-type GaAs pillar height is 800 nm, and the pillar surface diameter is 1200 nm (the sidewall has the inclination angle of 4.8° and the bottom diameter is 1100 nm). With these cavity parameters, we worked on the FEM simulation of the high-Q cavity mode. A resonance mode with a high-Q value was found at the wavelength of 1001.2 nm, which is very close to the measured resonance wavelength of 999.6 nm. It is the WG mode with the mode number of 9 (the WG mode in Fig. 3(a) has the mode number of 5) and the calculated cavity Q was about 5500. The observed Q value is lower than this simulated value and this shows that the observed Q value may be increased with the further improvement of the fabrication processes. The threshold gain for lasing operation is known to be inversely proportional to the cavity Q value[22] and is expected to be lower for the higher Q value.

In summary, we showed that the cavity resonance modes are probed employing the broad-band room temperature mid-gap emission of n-GaAs. With this method, we demonstrated the observation of the high cavity Q value of 3800 at room temperature, which is the extremely high Q value among metallic nanocavities. The FEM simulation predicted the higher Q value, and the further improvement of the fabrication processes may lead to the higher cavity Q value. We also found the general tendency that the higher-Q resonance modes are more frequently observed for the larger pillar diameter, and we explained this tendency with the FEM simulation. With the implementation of this high-Q resonance mode into nanolasers, lowering of the lasing threshold will be expected. If the compatibility of the cavity modes with the reported high photon extraction efficiency from the top air/semiconductor interface becomes possible in the metallic nanocavities, it will enable us to realize highly efficient photon coupling to cavity resonance modes and single-photon-based functional nanophotonic devices.

[1]T. H. Maiman, Nature **187**, 493 (1960).
[2]D. A. B. Miller, Proc. IEEE **97**, 1166 (2009).
[3]L. Pan and D. B. Bogy, Nat. Photonics **3**, 189 (2009).
[4]M. Loncar, A. Scherer, and Y. Qiu, Appl. Phys. Lett. **82**, 4648 (2003).

[5]Y. Nakayama, P. J. Pauzauskie, A. Radenovic, R. M. Onorato, R. J. Saykally, J. Liphardt, and P. Yang, Nature **447**, 1098 (2007).
[6]M. T. Hill, Y.-S. Oei, B. Smalbrugge, Y. Zhu, T. de Vries, P. J. van Veldhoven, F. W. M. van Otten, T. J. Eijkemans, J. P. Turkiewicz, H. de Waardt, E. J. Geluk, S.-H. Kwon, Y.-H. Lee, R. Notzel, and M. K. Smit, Nat. Photonics **1**, 589 (2007).
[7]C.-Y. Lu, S.-W. Chang, S.-H. Yang, and S. L. Chuang, Appl. Phys. Lett. **95**, 233507 (2009).
[8]M. T. Hill, M. Marell, E. S. P. Leong, B. Smalbrugge, Y. Zhu, M. Sun, P. J. van Veldhoven, E. J. Geluk, F. Karouta, Y.-S. Oei, R. Notzel, C.-Z. Ning, and M. K. Smit, Opt. Express **17**, 11107 (2009).
[9]M. P. Nezhad, A. Simic, O. Bondarenko, B. Slutsky, A. Mizrahi, L. Feng, V. Lomakin, and Y. Fainman, Nat. Photonics **4**, 395 (2010).
[10]C.-Y. Lu, S.-W. Chang, S. L. Chuang, T. D. Germann, and D. Bimberg, Appl. Phys. Lett. **96**, 251101 (2010).
[11]J. H. Lee, M. Khajavikhan, A. Simic, Q. Gu, O. Bondarenko, B. Slutsky, M. P. Nezhad, and Y. Fainman, Opt. Express **19**, 21524 (2011).
[12]C.-Y. Lu, S.-W. Chang, S. L. Chuang, T. D. Germann, U. W. Pohl, and D. Bimberg, Semicond. Sci. Technol. **26**, 014012 (2011).
[13]M. Khajavikhan, A. Simic, M. Katz, J. H. Lee, B. Slutsky, A. Mizrahi, V. Lomakin, and Y. Fainman, Nature **482**, 204 (2012).
[14]C. Symonds, G. Lheureux, J. P. Hugonin, J. J. Greffet, J. Laverdant, G. Brucoli, A. Lemaitre, P. Senellart, and J. Bellessa, Nano Lett. **13**, 3179 (2013).
[15]K. Ding, M. T. Hill, Z. C. Liu, L. J. Yin, P. J. van Veldhoven, and C. Z. Ning, Opt. Express **21**, 4728 (2013).
[16]K. Ding, L. Yin, M. T. Hill, Z. Liu, P. J. van Veldhoven, and C. Z. Ning, Appl. Phys. Lett. **102**, 041110 (2013).
[17]K. Yu, A. Lakhani, and M. C. Wu, Opt. Express **18**, 8790 (2010).
[18]H. Kumano, H. Nakajima, H. Iijima, S. Odashima, Y. Matsuo, K. Ijiro, and I. Suemune, Appl. Phys. Express **6**, 062801 (2013).
[19]H. Nakajima, H. Kumano, H. Iijima, and I. Suemune, Appl. Phys. Lett. **101**, 161107 (2012).
[20]A. A. Quivy, A. L. Sperandio, E. C. F. da Silva, and J. R. Leite, J. Cryst. Growth **206**, 171 (1999).
[21]A. D. Rakic, A. B. Djurisic, J. M. Elazar, and M. L. Majewski, Appl. Opt. **37**, 5271 (1998).
[22]G. Bjork, H. Heitmann, and Y. Yamamoto, Phys. Rev. A **47**, 4451 (1993).