Subwavelength metallic cavities with high-Q resonance modes

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Abstract

Metallic cavities have been extensively studied to realize small-volume nanocavities and nanolasers. However cavity-resonance quality (Q) factors of nanolasers observed up to now remain low (up to ~500) due to metal optical absorption. In this paper, we report the observation of highest Q factors of 9000 at low temperature and ~6000 near room temperature in a metallic cavity with a probe of sub-bandgap emission of Si-doped GaAs. We analyze the temperature dependence of cavity-mode resonance wavelengths and show that the refractive-index term dominates the measured temperature dependence. We also show that this refractive-index term is cavity-mode dependent and the fitting procedure offers a new method to identify cavity modes. We simulate the metallic cavity with finite-element method and attribute the high-Q cavity mode to a whispering gallery optical mode. This mode is shown to have isotropic polarization dependence of the output emission, which is preferable for quantum information applications.
1. Introduction

Metallic cavities are able to confine optical fields tightly inside cavities, and realization of nanolasers is expected with miniaturized cavity volumes. Room-temperature laser operations have already been reported with metallic-cavity nanolasers [1-6]. Threshold injection current is dependent on active volumes and it can be lower for smaller active volumes [7], and therefore nanolasers are advantageous to achieve extremely low threshold current operations. Efficiency of lasers is generally evaluated with optical gain necessary for laser operations, and high-performance nanolasers with lower threshold gain are possible with lower optical loss inside nanocavities. Optical loss in nanocavities is evaluated with cavity quality (Q) factors below threshold, but the Q factors measured on the reported nanolasers remain low below ~500 due to significant optical absorption loss of the constituent metals [1,3,4,6,8-11]. Much higher Q factors were simulated with surface plasmon polariton (SPP) nanocavities at low temperature, but they are highly dependent on environmental temperature and are substantially reduced at room temperature [12-15]. Most of the estimated Q factors were ~100. However, a high Q factor over 1000 was measured with SPP whispering gallery (WG) modes [16]. Simulations of metallic cavities were also performed, and the higher Q factors of ~1700 [17] and ~3400 [18] were expected. Recently we fabricated metallic cavities and observed the Q factor of 3800 [19]. Realization of higher Q factors leads to lower-threshold or thresholdless nanolasers by the reduced internal optical loss and also to realization of quantum electrodynamic (QED) devices.

In this paper, we study the detailed resonance properties of metallic nanocavities. We observe high cavity Q factors of 9000 at low temperature and ~6000 near room temperature (RT) with a probe of sub-bandgap emission of Si-doped GaAs. We derive a general analytical expression for the temperature dependence of a cavity resonance peak in a metallic cylindrical cavity and study measured temperature dependence of the cavity resonance mode. We work on finite-element method (FEM) simulations to identify the observed resonance modes and discuss how the high Q factors were made possible in our metallic nanocavity. We also study the polarization dependence of optical emission from the high-Q cavity mode.

2. Cavity preparation

The fabrication process of metallic cavities is shown in figure 1. Firstly we prepare pillar structures of different diameters ranging from 200 to 2000 nm on a GaAs (001) surface employing electron-beam (EB) lithography of hydrogen silsesquioxane (HSQ) resist and
subsequent inductively coupled plasma (ICP) reactive ion etching (RIE). The typical etching condition is the process pressure of 0.08 Pa, mixed gas flow of Cl₂ and argon (Ar) with the respective rate of 0.1 and 0.1 standard cc per minute (sccm), ICP and bias powers of both 100 W, and the substrate temperature of 50°C. This results in the etching rate of ~25 nm/min. The pillar whole surface is covered with 200-nm-thick SiO₂ dielectric layer with plasma-enhanced chemical-vapor deposition (PECVD). The example of scanning electron microscope (SEM) image is shown in figure 1(b), which was observed with the accelerating voltage of 5 kV. We can confirm uniform surface coverage of the pillar structures with transparent images under observations with the higher voltage of 30 kV. Then the surface is covered with ~1.5-µm-thick silver (Ag) with EB evaporation. We can expect high optical reflectivity over 97% in the wavelength range longer than 900 nm with Ag. After the pillars are embedded in the Ag layer, the Ag surface is pasted to a glass substrate with UV resin and the GaAs substrate is removed with mechanical polishing and the subsequent ICP-RIE. One of the completed pillar-surface SEM image after the substrate removal is shown in figure 1(e).

![Figure 1](image_url)

Figure 1. Metallic-cavity preparation process. (a) GaAs pillar prepared with ICP-RIE. (b) SiO₂ deposition with PECVD. (c) Cross-sectional schematic after Ag EB deposition. (d) Glass substrate is pasted on the Ag surface with UV resin, and the sample is turned over. The GaAs substrate is removed with mechanical polishing and subsequent ICP-RIE. (e) SEM image of completed metallic cavity surface.
3. Probe of cavity modes with sub-bandgap emission in doped GaAs

3.1. Sub-bandgap emission in doped GaAs

For the purpose of probing cavity modes, we studied the possibility of sub-bandgap emission in doped GaAs. Figure 2(a) shows the spectra measured on Si-doped and Zn-doped GaAs at 20 K. Although Zn is more heavily doped by 10 times, sub-bandgap emission is clearly observed only with the Si-doped GaAs. Si is amphoteric in GaAs and Si$_{Ga}$-Si$_{As}$ donor-acceptor-pair (DAP) emission has been recognized at ~1.49 eV (wavelength of 832 nm) with low doping concentration of ~10$^{15}$ cm$^{-3}$ [20,21]. With heavy doping up to 10$^{18}$~10$^{19}$ cm$^{-3}$, deep emission around 1.24 eV (1000 nm) was observed and was assigned to Si$_{Ga}$-V$_{Ga}$ complex [22]. However, the deep emission was later discussed from the viewpoint of Si$_{Ga}$-Si$_{As}$ DAP emission [23].

We examined the excitation-power dependence of the sub-bandgap emission of Si-doped GaAs, and both the broad peaks of ~1.37 eV and 1.18 eV in figure 2(a) showed the blue shift for the higher photo-excitation as is shown in figure 2(b) for the former peak. This is the general signature of the DAP emission [20]. These peaks are much lower in energy compared with the DAP emission peak of 1.48 eV reported for the lower doping level [20,21], but it is probable that the high Si doping and the resultant ionized Si atoms in GaAs (Si$_\text{Ga}^+$ and

4
Si\textsubscript{As}) attract each other through mutual long-range Coulomb interactions and form clusters of Si atoms in GaAs [23]. This generally results in the observed lower luminescence photon energy. The advantage of employing sub-bandgap emission for the probe of cavity modes is that photon-absorption of semiconductors is faint so that it does not significantly influence the cavity resonances.

3.2 Observation of high-Q resonance modes

Here we focus to the wavelength band of ~900 nm where we could observe high-Q cavity modes as described below. The expanded view of the photoluminescence (PL) spectrum of Si-doped GaAs is shown in the upper part of figure 3(a). It is almost featureless in this expanded view. The dimensions of the pillar mainly discussed here are as follows: The pillar surface diameter facing air is 1300 nm and the bottom surface diameter surrounded with SiO\textsubscript{2}/Ag is 1100 nm. Therefore this pillar has the average sidewall taper angle of 8.1° with the

\[\text{Figure 3.} \quad \text{(a) PL spectra measured at 20 K with as-grown Si-doped GaAs without cavity and with the metal-embedded cavity. (b) Lorentz function fit (solid line) of one of the sharp peaks observed in (a) (shown by open circles). (c) Excitation-power dependence of the PL FWHM of the sharp peak. (d) Temperature dependence of the PL FWHM of the sharp peak.}\]
height of 700 nm. With this Ag-embedded structure, we observed sharp PL peaks as shown in the lower part of figure 3(a). For this measurement, a He-Ne laser at the wavelength of 632.8 nm was focused to the target pillar employing an objective lens with the numerical aperture of 0.4. Luminescence from the Si-doped GaAs pillar was collected with the same objective lens and was dispersed with a double monochromator with the focal length of 500 mm. We observed similar sharp peaks from many other pillars with different diameters and different heights. Since no exciton emission is possible far below the GaAs energy gap, these sharp peaks are identified as the luminescence enhanced by cavity modes.

The observed sharp peaks are well fitted with the Lorentzian lineshape function and one example is shown in figure 3(b). From the full width at half maximum (FWHM), the resonance mode is found to have the high cavity Q factor of 9000. This is an extremely high Q factor ever observed among metallic nanocavities. We measured the excitation-power dependence of this resonance peak. As is shown in figure 3(c), PL FWHM remains almost constant against the increase of the excitation power. The temperature dependence of the PL FWHM is also shown in figure 3(d). The measurement error increases at the higher temperature due to the decrease of the PL intensity, but the property remains essentially the same at the higher temperature. These results also confirm that the observed sharp peak is the cavity mode. The Q factor corresponding to the measured FWHM near RT is ~6000 and is not much sensitive to the environmental temperature.

4. Temperature dependence of cavity resonance energy

4.1 Analysis

We attributed the observed sharp luminescence peak to the cavity mode of the metallic nanostructure from the above measurements. Cavity modes generally show much reduced temperature dependence of their resonance energies. However the temperature dependence of metallic cavities based on semiconductor nanostructures has not been quantitatively examined. We analyze resonance modes in a cylindrical metallic cavity and study the temperature dependence of the resonance-mode energy, $E$. The details of the analysis are presented in the appendix, and the finally derived formula for the temperature dependence is given as follows:

$$\frac{1}{E} \frac{dE}{dT} = -\frac{1}{\lambda} \frac{d\lambda}{dT} = -\alpha(T) - \zeta_m \beta(T) = -\mu(T), \quad (1)$$

where $\lambda$ is the resonance wavelength. $\alpha(T) = (1/R)(\partial R/\partial T)$ and $\beta(T) = (1/n_1)(\partial n_1/\partial T)$ are relative temperature dependences of linear thermal expansion coefficient and of refractive
index, respectively, where $R$ and $n_1$ are the radius and the refractive index of the pillar in the metallic cavity. $\zeta_{m,s} = (1 + f_{m,s})/(1 - g_{m,s})$ and the explicit expressions for $f_{m,s}$ and $g_{m,s}$ are given in the appendix, which are functions composed of the m-th-order Bessel function of the first kind and the Hankel function. $s$ is given as TE or TM for transverse-electric (TE) and transverse-magnetic (TM) modes, respectively. For WG modes, both $f_{m,s}$ and $g_{m,s}$ turn out to be zero as is discussed in the appendix, that is, $\zeta_{m,WG} = 1$. One example of numerically calculated results for the $\zeta_{m,s}$ factor is shown in figure 4 for $m=0$ and $s=TE$ as a function of the refractive index $n_1$ of the GaAs pillar. Except for some anomalies at specific refractive-index values, the $\zeta_{m,s}$ factor is close to one but shows gentle dependence on the GaAs refractive index.

Employing equation (1), the temperature dependence of the resonance-peak photon energy is given by,

$$E(T) = E(T_L) \exp \left[ \int_{T_L}^T -h(T)dT \right],$$

where $T_L$ is the lowest temperature of the measurements. The measured temperature dependence of the sharp peak is compared with the above analysis employing the measured values of $\alpha(T)$ and $\beta(T)$ for GaAs [24].

![Figure 4](image_url)

Figure 4. One example of numerically calculated results on the $\zeta_{m,s}$ factor. The parameters for the calculation are $R=600$ nm, $m=0$, $s=TE$, and the wavelength of 900 nm.
4.2 Temperature dependence measurements and discussion on cavity modes

The temperature dependences of the sharp peak energy and the PL peak observed with the Zn-doped GaAs sample are shown in figure 5 for comparison. As is shown in figure 2(a), the Si-doped GaAs PL peak is broadened due to electron filling in the conduction-band edge with the high n-type doping and this influence the measurement of the bandgap temperature dependence. The Zn-doped GaAs also broadens the band-edge luminescence with hole filling due to the p-type doping, but the higher density of states in the valence band shows up clearer band-edge PL peak, which is more convenient for the measurement. Due to this doping effect, the temperature dependence of the Zn-doped GaAs PL peak (blue circles) shows some anomalous behavior in the low-temperature range, but the overall temperature dependence is well fitted with the well-known Varshni relation (blue solid line) employing the reported physical parameters [25].

The measured cavity mode (red circles) showed much reduced temperature dependence. The four solid lines were calculated to fit the measured cavity mode by setting the $\xi_{m,s}$ factor in equation (1) to 1.0, 1.1, 1.2, and 1.3 considering the result shown in figure 4. The setting of $\xi_{m,s} = 1.0$ corresponding to WG modes well reproduces the measured temperature dependence.

![Graph](image_url)

**Figure 5.** Measured temperature dependence of the GaAs (Zn-doped) PL peak (blue circles) and the sharp cavity-mode peak observed in figure 3(b) (red circles). The blue solid line for GaAs is the plot of the Varshni relation and the four solid lines for the cavity mode are calculated with equations (1) and (2) by setting the $\xi_{m,s}$ factor to 1.0, 1.1, 1.2, and 1.3.
of the cavity mode. Although the deviations from the line for \( \zeta_{m,s} = 1.0 \) with the larger \( \zeta_{m,s} \) values are modest, this shows that the observed high-Q cavity mode is most probably the WG mode.

5. Cavity-mode simulation and discussion

We worked on FEM simulations to study the cavity modes and their resonance Q factors in more detail. The eigenmode solver of COMSOL was used to clarify the eigenstate of the cavity mode. Impedance boundary condition was set at the boundary between SiO\(_2\) and Ag. The refractive indices of GaAs and SiO\(_2\) were set to be 3.60 and 1.46, respectively. The permittivity of Ag was taken from the literature [26]. One of our simulation results is shown in figures 6(a) and 6(b), which are the electric field intensity distributions in the pillar cross-sections perpendicular and parallel to the height direction, respectively. This is a WG mode that has the mode number of 10 and the Q factor of 12240 at the resonance wavelength of 901.3 nm, which is close to the measured resonance wavelength shown in figure 3(b). This is the only one cavity mode found with the simulation of the given structure around the measured wavelength. The measured structure has the sidewall angle of 8.1° as described in section 3.2. We studied the influence of the sidewall angle by changing the bottom diameter while keeping the other parameters the same. We found a TE mode with the Q factor of 8500 at the wavelength of 905.2 nm near the measured wavelength with the sidewall angle of 3.3° (bottom diameter of 1220 nm). TM modes were not found in this series of simulations. Since the condition for the presence of the TE mode is beyond our structural measurement accuracy,

![Figure 6](image_url)

**Figure 6.** Electric-field intensity distributions of a WG mode in a cylindrical nanocavity calculated with FEM simulation. (a) Top view and (b) lateral view. The white arrows in (a) show the electric-field directions in the major intensity area (partially shown on the left half and the arrow size is modified depending on the field intensity). (c) is the calculated structure viewed from the same direction as (b). The dashed line shows the Ag/SiO\(_2\) interface outside the pillar area. The WG mode number is 10. The top and bottom surface diameters of 1300 nm and 1100 nm, respectively, and 700-nm of GaAs pillar height are assumed. The SiO\(_2\) thickness at the GaAs/Ag interface is 200 nm.
we conclude that the observed high-Q resonance mode is the WG mode. This is consistent with the mode assignment discussed in section 4.2.

Q factors in metallic nanocavities are limited by optical radiation loss and metallic absorption loss [27]. In SPP cavity modes, fields are highly confined to metal-dielectric interfaces and metallic absorption is generally more enhanced. Metal absorption loss is due to electron-phonon and electron-electron scattering in metals and is suppressed at low temperature [28]. Therefore Q factors of SPP cavity modes exhibit highly temperature-dependent properties [12-15]. Optical radiation loss occurs in dielectric cavities due to optical-field extension outside cavities [29]. Metal shield of dielectric cavities confines optical field toward inside of the cavities and reduces optical radiation loss [17,18]. Optical cavity modes show very low temperature dependence of Q factors, in contrast to the one of SPP cavity modes [14]. The weak temperature dependence of the Q factor observed in the present metallic cavity indicates that the metal absorption loss is reduced in our cavity. The higher Q factor observed in the present cavity in comparison to previous reports is attributed to the control of the SiO$_2$ thickness at the Ag/GaAs interface to reduce the metal absorption [18].

Isotropy of cavity-mode polarization is important to allow superposition of polarization states for a quantum emitter to transfer photon polarization qubits for quantum information applications [30]. We worked on the polarization measurements of the cavity modes and found that the high-Q cavity mode mainly discussed in this work exhibited nearly isotropic polarization of the optical emission as shown in figure 7(a). This isotropic property is

![Figure 7. Polar plot of the emission intensity measured on cavity modes as a function of the angle $\phi$ of a linear polarizer. (a) Cavity mode with Q=9000. (b) Cavity mode with Q=4500. (c) Cavity mode with Q=2000.](image-url)
consistent with the WG mode, of which electric field distribution has azimuthal symmetry as shown in figure 6(a). Additionally, we found a tendency that cavity modes with lower Q factors show properties approaching more linear polarizations as is exemplified by figure 7(b) for Q~4500 and figure 7(c) for Q~2000. The relation of near-field images and polarization of output emission has been studied by several groups. Even if azimuthally symmetric near-field images of cavity modes are observed, the polarization dependence of output emission can be angle-dependent [14,18,31,32]. It is conjectured that slight field localization induced by a slight structure asymmetry results in local field penetration toward metal-dielectric interfaces and lowers the cavity Q factors and that this also induces deviation from expected isotropic polarization dependence of the optical emission. Further study is necessary to reach definite conclusion on this relation.

6. Conclusion

We reported the observation of high Q factor of 9000 at low temperature and ~6000 near room temperature in metal-semiconductor nanocavities. We derived a general analytical formula for the temperature dependence of cavity-mode peak energy in a metallic cylindrical cavity and showed that the semiconductor-medium refractive-index term dominates the temperature dependence, which is dependent on cavity resonance modes. We showed that fitting of the measured temperature dependence with this analytical formula gives us a new method to identify cavity mode. Together with the FEM simulations of the metallic cavity, we attributed the high-Q cavity mode to a WG optical mode. We showed that this high-Q cavity mode exhibits isotropic polarization dependence of the output emission. For realizing QED devices with this kind of high-Q metallic nanocavities in near future, high photon input-output efficiencies are essentially important. Optical losses that limit the Q factors are the sum of metal absorption loss and photon radiation to outside cavities. In this regard, minimization of the metal absorption loss and efficient collection of photons emitted from the cavities satisfy this request. We have previously observed high external collection efficiency of ~18% photons generated from a quantum dot embedded in a similar metallic pillar structure [33], and the higher efficiencies are predicted by optimizing the structure [34]. Combination of this research with the present high-Q metallic cavity will open a way toward efficient QED devices in near future.
Appendix. Derivation of equations expressing the temperature dependence of resonance wavelength

In this appendix, we derive the expression for the temperature dependence of resonance wavelength in a cylindrical cavity. Let us consider a cylinder with radius \( R \) and refractive index \( n_1 \) surrounded by a material with refractive index \( n_2 \). We assume that the cylinder is infinite in \( z \) direction in the cylindrical coordinates. The cavity focused in the main text has two boundaries: one is the GaAs/SiO\(_2\) interface and the other is the SiO\(_2\)/Ag one. In the high Q mode mainly discussed in the text, the electromagnetic (EM) fields are well confined in the GaAs pillar. Therefore we ignore the outer boundary. In electrodynamics, it is well known that we can obtain eigen-modes of a cylindrical structure by solving the eigen-equation given below [35]:

\[
\begin{bmatrix}
\frac{1}{R \gamma_1} J'_m(R \gamma_1) - \frac{1}{R \gamma_2} H'_{m}^{(1)}(R \gamma_2) \\
\frac{n_1^2}{R \gamma_1} J'_m(R \gamma_1) - \frac{n_2^2}{R \gamma_2} H'_{m}^{(1)}(R \gamma_2)
\end{bmatrix}
\begin{bmatrix}
J_m(R \gamma_1) \\
H_m^{(1)}(R \gamma_2)
\end{bmatrix}
= \begin{bmatrix}
\frac{n k_z}{R^2 k_0} \\
\frac{1}{\gamma_1^2} - \frac{1}{\gamma_2^2}
\end{bmatrix}^2,
\]

where \( J_m \) and \( H_m^{(1)} \) are the Bessel function and Hankel function of the first kind with the order of \( m \), respectively. The prime denotes the derivation of the function. \( \gamma_1 \) and \( \gamma_2 \) are given by the expression

\[
\begin{align*}
\gamma_1^2 &= n_1^2 k_0^2 - k_z^2, \\
\gamma_2^2 &= n_2^2 k_0^2 - k_z^2,
\end{align*}
\]

where \( k_0 \) and \( k_z \) are wavevectors in vacuum and of the \( z \) component, respectively. As is clear from equation (A.1), the EM fields are generally not separable into TE and TM modes. For simplicity we consider the situation with no \( z \) component of the wavevector, where the EM fields are separated into TE and TM modes.

First let us consider the TM modes. The eigen-equation can be reduced to the form

\[
n_1 J'_m (n_1 k_0 R) H_m^{(1)}(n_2 k_0 R) = n_2 J'_m (n_1 k_0 R) H_m^{(1)}(n_2 k_0 R).
\]

Derivative of this equation with respect to temperature and rearrangement of the derivative terms result in
Namely and dividing both sides with equation (A.10), we obtain the following result:

\[
\left( n_1 J_m' H_m^{(1)} - \frac{n_2^2}{n_1} J_m' H_m^{(1)} \right) \frac{1}{k_0} \frac{\partial k_0}{\partial T} = - \left( n_1 J_m' H_m^{(1)} - \frac{n_2^2}{n_1} J_m' H_m^{(1)} \right) \frac{1}{R} \frac{\partial R}{\partial T}
\]

\[
- \left( n_1 J_m' H_m^{(1)} + \frac{1}{k_0 R} J_m' H_m^{(1)} - n_2 J_m' H_m^{(1)} \right) \frac{1}{n_1} \frac{\partial n_1}{\partial T},
\]

(A.5)

where we assumed that the refractive index \( n_2 \) is independent of temperature. Since \( k_0 = 2\pi/\lambda \), where \( \lambda \) is the wavelength, we obtain the relative temperature dependence of the eigen-wavelength as

\[
\frac{1}{\lambda} \frac{d\lambda}{dT} = - \frac{1}{k_0} \frac{\partial k_0}{\partial T} = \alpha(T) + \zeta_{m,s} \beta(T),
\]

(A.6)

where \( \alpha(T) = (1/R)(\partial R/\partial T) \) and \( \beta(T) = (1/n_1)(\partial n_1/\partial T) \). \( \zeta_{m,s} = (1 + f_{m,s})/(1 - g_{m,s}) \) and

\[
f_{m,s=\text{TM}} = \frac{1}{n_1} J_m' \left( \frac{1}{k_0 R} - n_2^2 \frac{H_m'(H_m')}{H_m''} \right), \quad g_{m,s=\text{TM}} = n_2^2 \frac{J_m'H_m'}{J_m''}, \quad n_1 \equiv n_1 J_m', \quad n_2 \equiv n_2 J_m'.
\]

(A.7)

In the case of TE modes, the eigen equation is reduced to the form:

\[
n_1 J_m (n_1 k_0 R) H_m'(n_2 k_0 R) = n_2 J_m (n_1 k_0 R) H_m'(n_2 k_0 R).
\]

(A.8)

Following the same procedure as the TM modes, we derive equation (A.6) with

\[
f_{m,s=\text{TE}} = \frac{1}{k_0 R} \frac{J_m'H_m'}{n_1 J_m'H_m' - n_2 J_m'H_m'} \quad \text{and} \quad g_{m,s=\text{TE}} = \frac{n_1^2 J_m'H_m' - n_2 J_m'H_m'}{n_1 J_m'H_m' - n_2 J_m'H_m'}.
\]

(A.9)

In what follows, let us focus to the case of a whispering gallery (WG) mode. The resonance condition of a WG mode is given by

\[
2\pi n_1 R = n\lambda,
\]

(A.10)

where \( n \) is an integer. Taking the derivative of equation (A.10) with respect to temperature and dividing both sides with equation (A.10), we obtain the following result:

\[
\frac{1}{\lambda} \frac{d\lambda}{dT} = \alpha(T) + \beta(T).
\]

(A.11)

Namely \( f_{m,s}=0, g_{m,s}=0, \) and \( \zeta_{m,s} = 1 \) for the WG mode.
References


The parameter $E(0)$ was shifted by 0.042 eV to the lower energy for the best fit to our measurements.

[27] Kwon S H, No Y S and Park H G 2014 Nano Convergence 1 8