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Characterization of Fabry-Pérot microcavity modes in GaAs nanowires fabricated by selective-area metal organic vapor phase epitaxy

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The authors present the formation of Fabry-Pérot cavity in single GaAs nanowire prepared by selective-area metal organic vapor phase epitaxy. The grown nanowires with hexagonal cross section are highly uniform and vertically oriented. Microphotoluminescence measurements of single GaAs nanowire exhibit periodic peaks in the intensity, which are suggestive of the longitudinal modes of a Fabry-Pérot cavity. The cavity is formed along the length of the nanowire and the (111) facets of both ends act as reflecting mirrors. Additionally, optical waveguides in GaAs nanowires were also observed. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2787895]

Semiconductor nanowires have been attracting great interests because they are potentially used as building blocks in nanoelectronics and photonics.[1-4] Recently, semiconductor nanolasers working in ultraviolet region based on ZnO (Ref. 3) and CdS(Ref. 4) single nanowires were reported. These applications require that the fabricated nanowires should be uniform and flat enough to form Fabry-Pérot cavities. We recently proposed and demonstrated an alternative method of preparing semiconductor nanowires by using selective-area metal organic vapor phase epitaxy (SA-MOVPE).[5-8] Compared to the conventional vapor-liquid-solid growth,[9,10] this is a catalyst-free approach and only relies on the thermal decomposition of source materials. Furthermore, since this method fully utilizes the nature of epitaxial growth, it is expected to exhibit superior crystalline quality as well as good controllability of the growth with atomic precision to form abrupt doping profiles and heterojunctions, including vertical and lateral heterostructures.[6-8]

In this letter, we fabricated GaAs nanowires with uniform and smooth surfaces by SA-MOVPE. Microphotoluminescence (μPL) measurements reveal the formation of Fabry-Pérot cavity modes in individual GaAs nanowires, caused by the refractive index difference between the wire and the surrounding. Thus, GaAs nanowires may be candidate materials for achieving stimulated emission or lasing, especially in near-infrared region.

SA-MOVPE growth of GaAs nanowires started with the deposition of 30 nm SiO_2 layer by plasma sputtering on (111)B GaAs substrate. Then SiO_2 was partially removed by electron beam lithography and wet chemical etching. The mask pattern of SiO_2 had circular opening and they were arranged in a triangular lattice. The diameter of the grown nanowires was directly related to that of the opening holes. In addition, we controlled the nanowire length by adjusting the opening pitch and growth condition. Finally, the patterned substrates were loaded into a horizontal low-pressure MOVPE system working at 0.1 atm using trimethylgallium (TMG) and arsine (AsH_3) as source materials. Typical partial pressures were 9.6×10^{-7} and 2.5×10^{-4} atm for TMG and AsH_3, respectively. The growth temperature and growth time

for GaAs nanowires were 750 °C and 60 min. Typical freestanding GaAs nanowires grown on (111)B GaAs substrate by MOVPE are illustrated in Fig. 1(a). Scanning electron microscopy (SEM) studies clearly indicated a uniform array of vertical standing GaAs nanowires on the substrate. The cross section of the nanowires is hexagonal and the top and side surfaces are very flat, indicating that they are surrounded by six $\{1\bar{1}0\}$ facets and the top (111)B facet.

Before μPL measurements, the GaAs nanowires were mechanically cut down and dispersed onto a SiO_2-coated Si substrate, on which Ti/Au metal markers had been defined by photolithography. This enabled us to locate individual wires and to perform optical measurements and SEM on the same nanowires. Then μPL measurements were carried out using a 632.8 nm continuous-wave He–Ne laser for excitation. The excitation beam was focused onto $\sim$2 μm in diameter with a ×50 microscope objective on the sample placed in a cryostat, giving a power density of $\sim$1050 W/cm^2. The excitation power was adjusted using different combinations of neutral density filters. The emission through the same microscope objective was collected into a liquid-N$_2$-cooled charge coupled device (CCD) for spectral analysis or a CCD camera for imaging. SEM images of a typical nanowire used for PL measurements in this study are shown in Fig. 1(b). The nanowire dispersed on Si substrate is well defined and extremely uniform. The length (L) is 4.5 μm and the diameter (D) is 320 nm.
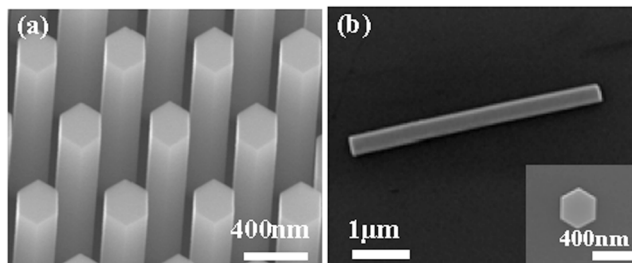

FIG. 1. (a) Typical Bird's eye SEM image of freestanding GaAs nanowires fabricated by SA-MOVPE. (b) Top view SEM image of single GaAs nanowire used for optical measurements, inset shows hexagonal cross section of this nanowire.

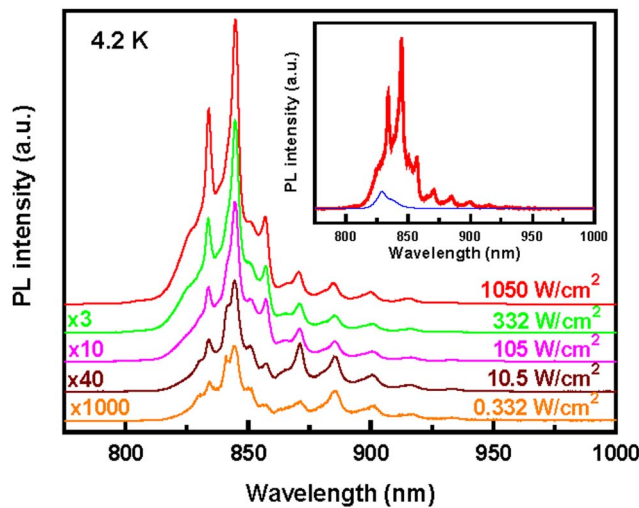[a]Author to whom correspondence should be addressed. Electronic mail: hua@rciqe.hokudai.ac.jp

FIG. 2. (Color online) 4.2 K PL spectra of a 5.0-μm-long GaAs nanowire recorded at excitation power densities of 1050, 332, 105, 10.5, and 0.332 W/cm^2, respectively. Inset shows that the 4.2 K PL intensity of the nanowire with cavity modes (upper) is ten times stronger than that of the nanowire without cavity modes (lower).

4.2 K PL spectra of a GaAs nanowire (L=5.0 μm, D=300 nm) with different excitation power densities are illustrated in Fig. 2. The excitation power density was changed stepwise from 1050 to 0.33 W/cm^2. Usually, the emission of GaAs nanowires is very weak due to nonradiative recombination of photoexcited carriers at the air-exposed GaAs sidewall surface where high density of surface states exists.[6] But in our study, some single nanowires exhibit relatively stronger emission in near-infrared region. Interestingly, a series of periodic peaks is observed in PL spectra ranging from 830 to 940 nm and the excitation power densities have no influence on the peak wavelengths. These distinct peaks should correspond to the resonant peak of different longitudinal modes of Fabry-Pérot cavity in GaAs nanowire and the peak wavelength is the mode wavelength. In principle, if the ends of the nanowire are smooth enough to function as two reflecting mirrors, the nanowire can define a Fabry-Pérot optical cavity with modes $m=2nL/\lambda$,[4] where m is an integer, L is the length of the cavity, λ is the wavelength, and n is the refractive index. For comparison, 4.2 K PL spectrum of another nanowire (L=4.3 μm, D=190 nm) without Fabry-Pérot cavity modes is presented in the inset of Fig. 2. The PL intensity of GaAs nanowire with cavity modes is about ten times stronger than that of the nanowire without cavity modes. It is observed that the nanowires with a diameter less than 200 nm are difficult to form Fabry-Pérot microcavities. One reason is that the larger diameter will help in enhancing the optical confinement and decreasing the optical losses effectively.[11] Additionally, the uniform thickness and smooth surface morphology of the nanowire are essential in this phenomenon because the nonuniformity of the nanowire leads to large optical losses from surface emission.

Figure 3(a) exhibits temperature-dependent PL spectra of the 5.0-μm-long GaAs nanowire. The PL spectra were recorded at 4.2, 50, 100, 150, and 200 K respectively. As temperature increases, the emission band at the center of about 830 nm becomes dominant and the peak linewidth broadens. This emission peak is attributed to the band edge of GaAs emission.[12] The redshift of this peak is nearly 25 nm with the temperature from 4.2 up to 200 K. These experimental re-
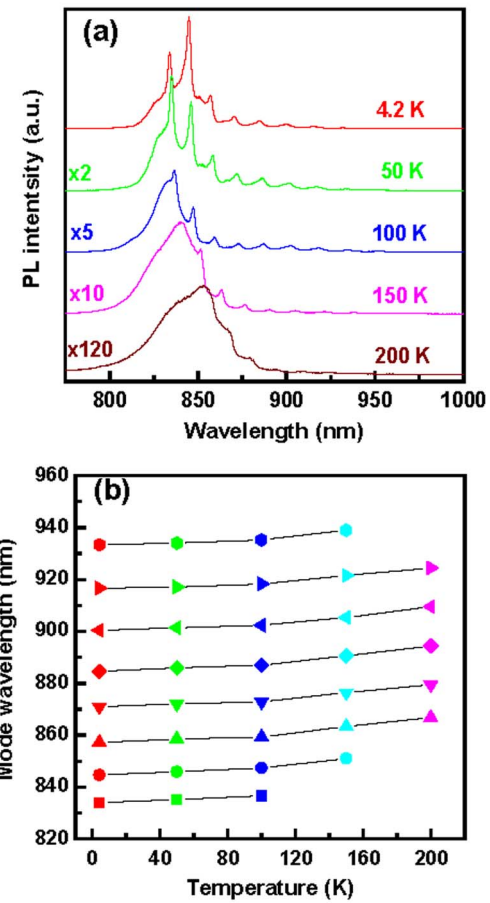

FIG. 3. (Color online) (a) Temperature-dependent PL spectra of a 5.0-μm-long GaAs nanowire with an excitation power density of 1050 W/cm^2. (b) Mode wavelengths of PL spectra (a) recorded at the temperatures of 4.2, 50, 100, 150, and 200 K, respectively.

sults are well consistent with the well-known Varshni formula. However, the wavelengths of cavity modes do not change so much with temperatures. Figure 3(b) clearly indicates the positions of the mode wavelengths from 830 to 940 nm with increasing temperature. Varying the temperature from 4.2 to 200 K, the mode wavelengths just redshift around 8–9 nm, which is much smaller than that assigned to the band edge. The reason is explained as follows. Based on the formula of $m=2nL/\lambda$, the mode wavelength of the nanowire is only affected by the material refractive index. With the temperature from 4.2 up to 200 K, the variation of the GaAs index is just at an order of magnitude of 0.01;[13] thus, the corresponding change of the mode wavelength is approximately 1% as well.

For Fabry-Pérot cavity modes, the mode spacing $\Delta\lambda$ for a cavity with length L is given by $\Delta\lambda=\lambda^2/\{2L[n-\lambda \times(dn/d\lambda)]\}$[4] where n is the refractive index and $dn/d\lambda$ is the first-order dispersion of the index. Group refractive index is defined as $n-\lambda(dn/d\lambda)$. This expression provides a good description of the observed mode spacing and the refractive index when the measured nanowire length is equated with L. Figure 4(a) shows 4.2 K PL spectra of the GaAs nanowires with different lengths. As expected, we obtain an inverse relationship between the mode spacing and nanowire length. According to these experimental results, the estimation of wavelength-dependent group refractive indices of GaAs nanowires is plotted in Fig. 4(b). For comparison, the wave-
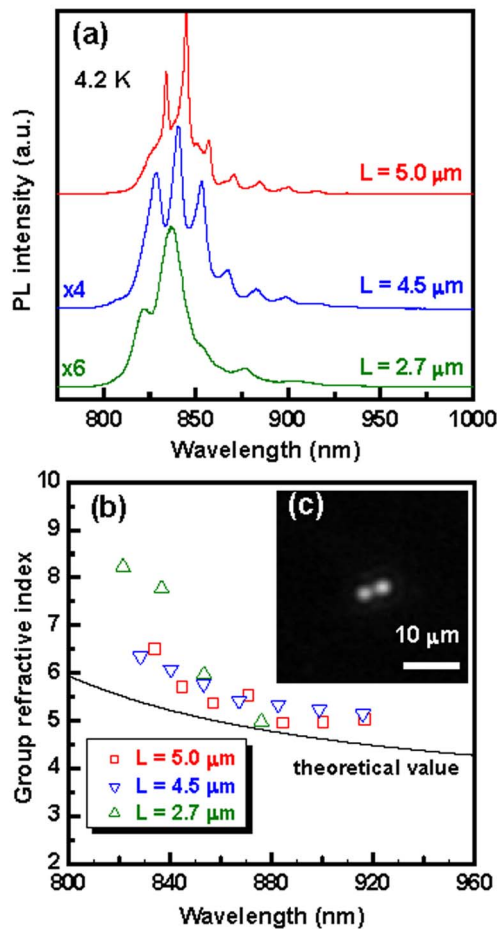
FIG. 4. (Color online) 4.2 K PL spectra of the GaAs nanowires with the lengths of 5.0, 4.5, and 2.7 μm (power density ~1050 W/cm^2). (b) Calculated group refractive indices of different length nanowires according to Fabry-Pérot cavity modes. The theoretical group index of GaAs is also given for comparison. (c) Optical image of a 4.5-μm-long GaAs nanowire excited by a He–Ne laser, indicating strong waveguiding properties.

length dependence of the theoretical index is also given, which was calculated from Sellmeier equation.[14] Here we neglect the temperature dependence on refractive index since its influence is negligible as mentioned previously. Most of the experimental data seem to agree with the theoretical tendency of the wavelength-dependent index except that the experimental values appear 0.2–1.1 higher than the theoretical ones. One possibility is due to the anisotropy of refractive index in GaAs. On the other hand, the indices of the 2.7-μm-long nanowire appear to disagree with those theoretical values. The mechanism needs further investigation, but we think that, in simple observation, relatively shorter nanowires cannot fit a Fabry-Pérot longitudinal mode so well, compared to those longer nanowires, because in this case the cavity length has been comparable with the mode wavelength. The similar phenomenon was also observed in ZnO nanoribbon microcavity.[15]

In addition, we can estimate the mode quality (Q) factor with different nanowires at 4.2 K, which is defined as the ratio of the mode wavelength to the width.[16] In Fig. 4(a), the width of the mode peak significantly decreases as the nanowire length increases. This indicates that the enhancement of the nanowire length can improve Q value efficiently. In the case of a 5.0-μm-long nanowire, the Q value at the wave-

length of 845 nm is estimated to be around 220 at 4.2 K. Compared to those nanowires reported to achieve lasing,[17,18] the Q value of the GaAs nanowires is low, indicating that the cavity loss is too large for lasing. For further optimization of the Q value, longer GaAs nanowires are strongly expected. Another feasible method is to fabricate core-shell nanowires instead of bare GaAs nanowires, e.g., GaAs/AlGaAs nanowires. It has been confirmed that the emission efficiency in core-shell heterostructures is significantly enhanced because the surrounding AlGaAs barrier prevents the coupling of photoexcited carriers in GaAs with the surface states, resulting in the high-quality and optically active nanowires.[6] Finally, during the spectra measurements of single GaAs nanowire, optical images were recorded with a laser filter when the excitation beam was focused at the center of the nanowire. As shown in Fig. 4(c), spontaneous emission was observed from both ends of GaAs nanowire, indicative of strong waveguiding properties in nanowire.

Although there are some scientific and technical challenges that need to be addressed such as the development of more efficient cavities to achieve higher Q value, our results may point to a possible route in the realization of stimulated emission or lasing within the wavelengths covering a near-infrared region, inside compound semiconductor nanowires.

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