Electrically driven lasing in light-emitting devices composed of n-ZnO and p-Si nanowires

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Abstract

Electrically driven lasing was demonstrated in light-emitting devices composed of n-ZnO and p-Si nanowires (NWs). The ZnO NWs were synthesized by thermal chemical vapor deposition and the Si NWs were formed by crystallographic wet etching of a Si wafer. The p–n heterojunction devices were constructed using the NWs by the direct transfer and dielectrophoresis methods. At an excitation current of 2 \(\mu\)A, the electroluminescence spectrum showed lasing behavior, and this phenomenon was explained by the ZnO-nanostructure-related cavity property.

(Some figures in this article are in colour only in the electronic version)

Electrically driven lasers have attracted much attention owing to the rapid advances made in the field of light-emitting devices. Recently, many studies have been conducted on devices constructed with various nanomaterials [1–7]. Among the various kinds of nanomaterials, ZnO is a promising material for optoelectronic devices due to its large binding energy of excitons and direct band gap, leading to an effective emission at room temperature. Besides, its one-dimensional (1D) structure is very useful for stimulating light as an optical cavity. Based on the optical and structural characteristics, ZnO nanowires (NWs) have been applied to various optoelectronic devices such as photodetectors, light-emitting diodes, and lasers [1–3, 8, 9].

Many attempts have been made to induce lasing behavior in ZnO NW-based devices, and most of these have focused on optically pumped lasers [10–13]. However, these lasers have several restrictions such as laser source and high power threshold [1]. Accordingly, electrically driven lasing in ZnO NWs has been of great interest [2, 14–16]. In this study, the electrically driven lasing behavior was demonstrated in the p–n heterojunction devices constructed using n-ZnO and p-Si NWs through a novel fabrication route.

The ZnO NWs were synthesized by thermal chemical vapor deposition on an Au-deposited SiO\(_2\)/Si substrate. The ZnO powder was ground in a ball-milling machine and then thermally evaporated at 1380°C under Ar atmosphere. The as-grown ZnO NWs exhibited an average diameter and length of \(~100\) nm and \(~12\) \(\mu\)m, respectively. The growth direction was perpendicular to the \((1\bar{1}2\bar{0})\) planes, and the NW-end shape was somewhat rounded [17]. The ZnO NWs were dispersed in methanol solution to align them during the device-fabrication process. The Si NWs were formed by crystallographic wet etching of a boron-doped (100)-oriented Si wafer with a doping concentration of \(~10^{20}\) cm\(^{-3}\). The etching was conducted using a 20 wt% potassium hydroxide (KOH) solution at 40°C.

The ZnO NWs were aligned by a dielectrophoresis (DEP) force under AC bias with an amplitude of 10 V at 1 kHz. The DEP force means the forces exerted on the dielectrics are subjected to nonuniform electric field. The electrical characteristics were examined by a semiconductor parameter analyzer (4155C, Agilent). The light-emission image was acquired by a charge-coupled device (CCD: 1612C, Scion). The electroluminescence (EL) spectra were analyzed using a spectrophotometer (USB2000plus, Ocean Optics) at room temperature in the dark. The contact property of crossed NWs at the p–n heterojunction was observed by scanning electron microscopy (SEM: S-4300, Hitachi).

The entire fabrication processes of the light-emitting devices composed of n-ZnO and p-Si NWs are shown in figure 1. The active regions for a Si NW are patterned by a
Figure 1. Fabrication processes of the light-emitting device composed of n-ZnO and p-Si NWs. (a) Formation of Si lines (450 nm (W) × 600 nm (H) × 170 μm (L)) by ICP etching. (b) Formation of an inverted-triangular-shaped Si NW by crystallographic wet etching. (c) Transfer of the Si NW onto a SiO2/Si substrate by a direct transfer method. (d) Deposition of Au/Ni and Al electrodes. (e) Alignment of the ZnO NW by a DEP technique. (f) Deposition of Au/Al electrodes.

A photolithography process, which is followed by the formation of Si lines (450 nm (W) × 600 nm (H) × 170 μm (L)) by inductively coupled plasma (ICP) etching (figure 1(a)). Crystallographic wet etching is conducted in order to shape an inverted-triangular-shaped Si NW with a base length of ∼100 nm, which is attributed to the large etching velocity of (111) surfaces (figure 1(b)) [18]. The Si NW is then transferred onto a SiO2/Si substrate by a direct transfer method (figure 1(c)). To prepare contact pads of the Si NW, two end positions of the Si NW are patterned by photolithography, and native oxide in the patterned region is removed by a HF solution. Then, Au/Ni is evaporated, and the lift-off processes are conducted. Also, Al electrodes are prepared at certain positions to define the position of the single ZnO NW, through the same processes such as photolithography, evaporation, and lift-off. Figure 1(d) represents the positions of patterned metal electrodes. Subsequently, the ZnO NW-dispersed solution is dropped between the Al electrodes. When AC electric field is applied, the ZnO NW is aligned between the electrodes by a DEP force, and forms the p–n heterojunction with the p-Si NW (figure 1(e)). The DEP force (F_DEP) that makes use of the polarization of the ZnO NW is represented by the following equation,

$$F_{\text{DEP}} = \frac{v}{2} \epsilon_m \alpha_r \nabla (E_{\text{RMS}}^2),$$

where $v$ is the volume of the NW, $\epsilon_m$ is the permittivity of the medium, $\alpha_r$ is the real part of the Clausius–Mosotti factor, and $E_{\text{RMS}}$ is the root-mean-square (RMS) value of the AC electric field [19]. Finally, one end of the aligned ZnO NW is capped with the first electrode of Au/Al and a part of the ZnO NW is connected to the second electrode (figure 1(f)). Therefore, the other end of the ZnO NW remains open (figure 1(f)). Figure 2(a) represents the optical image of the crossed NW p–n heterojunction. Figure 2(b) shows the SEM image of the p–n heterojunction constructed with the crossed NWs, confirming that both of the NWs are well contacted.

Figure 3 shows the CCD image of the light propagation along the single ZnO NW. The arrow indicates the location of the incident laser focus. The wavelength of the laser used here is 457 nm, the energy of which is below the band gap of ZnO. Therefore, the strength of the photoluminescence is expected to be very low and we could observe the light of the incident laser focus. The wavelength of the laser used here is 457 nm, the energy of which is below the band gap of ZnO. Therefore, the strength of the photoluminescence is expected to be very low and we could observe the light...
The lasing modes of luminescence by the band-to-band transition. The large deviation of \( \eta \) was estimated to be \( \sim 24 \) from the slope of the \( \ln(I) \) versus \( V \) plot, as shown in the inset of figure 4(a), by the equation \( \eta = e/k_B T [\beta (\ln I)/\delta V]^{-1} \), where \( k_B \) is a Boltzmann constant and \( T \) is the operating temperature. The large deviation of \( \eta \) from the ideal case (\( \eta = 1 \), thermionic emission model) may be attributed to the existence of an unavoidable interfacial layer during the fabrication processes such as patterning and/or dielectrophoresis steps [22, 23]. Figure 4(b) shows the EL spectrum at the excitation current of 2 \( \mu \)A. The strong two peaks at the blue region are assigned to the resonant wavelengths \( \lambda_{1} \approx 386 \) nm and \( \sim 394 \) nm, respectively. (The peak of \( \sim 415 \) nm was not fitted well due to the slope of the background.) The relatively low signal/noise ratio in our EL spectrum may be attributed to the rounded end shape of the used ZnO NW. The cavity-end flatness is one of the important parameters because well-defined faceting ends enable unique optical confinement, leading to low lasing threshold and high lasing gain [24]. Unfortunately, it was difficult to construct the heterojunction device with various ZnO nanostructures due to the complexity of our fabrication processes. The longitudinal mode spacing (\( \Delta \lambda \)) of a Fabry–Pérot cavity is given by the equation,

\[
\Delta \lambda = \frac{L}{L} \left[ \frac{\lambda^2}{2} \left( n - \frac{dn}{d\lambda} \right) \right]^{-1},
\]

where \( L \) is the length of the cavity, \( n \) is the refractive index of the ZnO NW (\( = 2.45 \)), \( \lambda \) is the resonant wavelength (\( \approx 394 \) nm), and \( dn/d\lambda = -0.015 \) nm\(^{-1} \) [25, 26]. On the assumption that two peaks are primarily axial modes, the

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**Figure 3.** CCD image of light propagation along the ZnO NW. The arrow indicates the location of incident laser focus. The inset shows the as-prepared ZnO NW.

**Figure 4.** (a) \( I-V \) curve obtained from the p–n heterojunction device, showing the turn-on voltage of about 3.6 V and the breakdown voltage of below \( -30 \) V. The inset shows the \( \ln(I) \) versus \( V \) plot to acquire the diode-ideality factor from the slope of the fitting curve. (b) EL spectrum of the p–n heterojunction device at the excitation current of 2 \( \mu \)A, with the fitting results using five Gaussian functions. The lasing peaks are observed at \( \sim 386 \) and \( \sim 394 \) nm. The inset shows the CCD image of the electrically driven lasing. (c) The cross-sectional schematic view of the p–n heterojunction device.
mode spacing of ~8 nm is matched with the cavity length of ~1.2 μm. If it is the case, this spectrum cannot be explained as pure axial modes, considering the length of our ZnO NW (~12 μm). However, the presence of two peaks at 386 and 394 nm indicates the clear evidence on stimulated emission that cannot be acquired by spontaneous emission. The optically pumped lasing characteristics of well-faceted ZnO nanoribbons were reported by Yang’s group, with various nanoribbon lengths in the range from 4.5 to 18 μm [24]. The ZnO nanoribbon of 18 μm clearly exhibited longitudinal modes with sharp line widths, and the decrease in the luminescence-peak number and the broadening of line widths were observed with the decrease in the ZnO-nanoribbon length due to insufficient gain for primarily axial modes. The luminescence spectra, showing two relatively broad peaks as a beginning regime of stimulated emission, illustrate the close similarity between our data (ZnO nanowire, ~12 μm) and Yang’s group’s data (ZnO nanoribbon, 4.5–10 μm), and it is thought that the small difference in the length-dependent regime is reasonable, considering the nanostructural differences such as geometry, growth direction, and faceting degree. Actually, with 1D nanostructures, the lasing physics at the beginning of the regime of stimulated emission has not been fully elucidated until now, even in other groups. Therefore, it would be more meaningful to accurately control ZnO nanostructures, and thereby correlate these nanostructures with the lasing properties of the device in the entire lasing regime. The cross-sectional schematic view of the light-emitting device is presented in figure 4(c). The holes injected from the p-Si NW and the electrons in the n-ZnO NW are recombined to photons at the crosspoint. Because the location producing photons is limited, the light is coupled to the NW as a guided mode propagating along the NW after being sufficiently accumulated in the ZnO-NW cavity. Eventually, the stimulated photons are emitted through the open side of the ZnO NW, as shown in the inset of figure 4(b).

In conclusion, electrically driven lasing in light-emitting devices composed of n-ZnO and p-Si NWs was achieved in this work. The ZnO and Si NWs were used to implement a p–n heterojunction light-emitting device by the direct transfer and DEP methods. In the EL spectrum, the lasing peaks were observed at 386 and 394 nm, and this phenomenon was explained by the ZnO-nanostructure-related cavity property. Further studies are needed to improve the device performance with accurate nanostructure control and better understanding of lasing physics.

Acknowledgments

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References