Ultraviolet Lasing From Vertically-Aligned ZnO Nanowall Array

Kyushu University in Japan
Masahiro Takahashi, Kosuke Harada, Shihomi Nakao,
Mitsuhiro Higashihata, Hiroshi Ikenoue,
Daisuke Nakamura, Tatsuo Okada
Zinc Oxide

Characteristics of ZnO

- Wide band gap energy: 3.37 [eV]
- Large exciton binding energy: 60 [meV]
- Harmless
- A low price (about 2$/kg)

In addition...

- ZnO is synthesized nanocrystals easily.
- Nanocrystals have high crystallinity.

Expect to apply to light-emitting devices in UV region.
A lot of ultraviolet lasing are reported because ZnO nanostructures have high-crystallinity.

- **Micro cavity lasing**
  - Lasing from single crystal like a ZnO nanowire.
  - This is because internal reflection due to the high refractive index of ZnO.

  - Fabry-Perot (FP) cavity
  - Whispering-gallery mode (WGM) cavity

- **Random lasing**
  - Lasing from complicated oscillation routes randomly formed by a number of ZnO nanocrystals or a single ZnO micrometer-sized random medium.
A lot of ultraviolet lasing are reported because ZnO nanostructures have high-crystallinity.

- Micro cavity lasing
  - Lasing from single crystal like a ZnO nanowire.
  - This is because internal reflection due to the high refractive index of ZnO.
    - Fabry-Perot (FP) cavity
    - Whispering-gallery mode (WGM) cavity

- Random lasing
  - Lasing from complicated oscillation routes randomly formed by a number of ZnO nanocrystals or a single ZnO micrometer-sized random medium.

**Purpose**
We have succeeded in synthesizing ZnO nanostructures.
In this study, we investigated their optical characteristics.
Steps to synthesize ZnO nanostructure

1. Depositing ZnO buffer layer on a sapphire substrate
2. Making patterning using two beam laser interference irradiation to the buffer layer.
3. Fabricating ZnO nanostructures by NAPLD

**Experimental conditions**
- Ablation laser: 3rd harmonic of Q-switched Nd:YAG laser (1.5 J/cm²)
- Substrate temperature: 500 °C
- Gas pressure: 3.0 Pa (2.25 X 10² torr)
- Deposition time: 10 min
- Atmosphere gas: O₂, 5 sccm
1. Depositing ZnO buffer layer on a sapphire substrate
2. Making patterning using two beam laser interference irradiation to the buffer layer.
3. Fabricating ZnO nanostructures by NAPLD

Experimental conditions
Ablation laser: 3rd harmonic of Q-switched Nd:YAG laser (1.5 J/cm²)
Substrate temperature: 500 °C
Gas pressure: 3.0 Pa (2.25 X 10⁻² torr)
Deposition time: 10 min
Atmosphere gas: O₂, 5 sccm
Steps to synthesize ZnO nanostructure

1. Depositing ZnO buffer layer on a sapphire substrate
2. Making patterning using two beam laser interference irradiation to the buffer layer.
3. Fabricating ZnO nanostructures by NAPLD

**Experimental conditions**

- Ablation laser: 3rd harmonic of Q-switched Nd:YAG laser (1.5 J/cm²)
- Substrate temperature: 500 °C
- Gas pressure: 3.0 Pa (2.25 X 10⁻² torr)
- Deposition time: 10 min
- Atmosphere gas: O₂, 5 sccm
Steps to synthesize ZnO nanostructure

1. Depositing ZnO buffer layer on a sapphire substrate
2. Making patterning using two beam laser interference irradiation to the buffer layer.
3. Fabricating ZnO nanostructures by NAPLD

Experimental conditions

- Ablation laser: 3rd harmonic of Q-switched Nd:YAG laser (1.5 J/cm²)
- Substrate temperature: 500 °C
- Gas pressure: 3.0 Pa (2.25 × 10⁻² torr)
- Deposition time: 10 min
- Atmosphere gas: O₂, 5 sccm
1. Depositing ZnO buffer layer on a sapphire substrate
2. Making patterning using two beam laser interference irradiation to the buffer layer.
3. Fabricating ZnO nanostructures by NAPLD
The periodic pattern can be realized in a single shot.
Fabricating ZnO nanostructures
NAPLD method

1. Depositing ZnO buffer layer on a sapphire substrate
2. Making patterning using two beam laser interference irradiation to the buffer layer.
3. Fabricating ZnO nanostructures by NAPLD
Fabricating ZnO nanostructures

NAPLD method

**Experimental conditions**

Ablation laser: 3rd harmonic of Q-switched Nd:YAG laser (1.0 J/cm²)

Substrate temperature: 750 °C

Gas pressure: 200 Torr

Deposition time: 60 min

Atmosphere gas: Ar, 20 sccm

---

Before deposition

2.0 μm

60 min, 0°

60 min, 60°
Fabricating ZnO nanostructures

NAPLD method

Experimental conditions

Ablation laser: 3rd harmonic of Q-switched Nd:YAG laser (1.0 J/cm²)
Substrate temperature: 750 °C
Gas pressure: 200 Torr
Deposition time: 60 min
Atmosphere gas: Ar, 20 sccm

ZnO nanowall
Height: about 5 μm
Thickness: about 180 nm

Before deposition

60 min, 0°

60 min, 60°
Optical characteristics measurement

- Excitation-cut laser beam (355 nm)
- Objective lens (X 50)
- ZnO nanostructure
- Sapphire substrate
- SiO₂ substrate
- Half mirror
- Light fiber
- Spectrometer
- CCD

 KYUSHU UNIVERSITY LASER LABORATORY
Lasing in a single piece of ZnO nanowall

threshold power density for lasing 150 kW/cm²
Lasing in a single piece of ZnO nanowall

The mode spacing ($\Delta \lambda$) is inversely proportional to cavity length ($L$).

\[
L = \frac{\lambda_m^2 - \lambda_m \Delta \lambda}{\Delta \lambda (n_m - \lambda_m \frac{dn}{d\lambda})}
\]
Lasing in a single piece of ZnO nanowall

The mode spacing ($\Delta \lambda$) is in inversely proportional to cavity length ($L$).

$$L = \frac{\lambda_m^2 - \lambda_m \Delta \lambda}{\Delta \lambda (n_m - \lambda_m \frac{dn}{d\lambda})}$$
Lasing in a single piece of ZnO nanowall

The mode spacing ($\Delta \lambda$) is inversely proportional to cavity length ($L$).

$$ L = \frac{\lambda_m^2 - \lambda_m \Delta \lambda}{\Delta \lambda (n_m - \lambda_m \frac{dn}{d\lambda})} $$

Many different oscillation routes are formed inside it.
Lasing from vertically-aligned ZnO nanowall

Intensity [arb. unit]

Wavelength [nm]

10 μm

CCD image

1.0 μm

SEM image

threshold power density for lasing 1150 kW/cm²

Excitation power [kW/cm²]

Intensity [arb. unit]

Excitation power [kW/cm²]

387.6 nm
Lasing from vertically-aligned ZnO nanowall is achieved for the first time.
Lasing from vertically-aligned ZnO nanowall

- ZnO nanowalls were excited from three different angles by rotating the sample in-plane.
Lasing from vertically-aligned ZnO nanowall

- ZnO nanowalls were excited from three different angles by rotating the sample in-plane.

![CCD image](image)

![Graph](graph)
Lasing from vertically-aligned ZnO nanowall

- ZnO nanowalls were excited from three different angles by rotating the sample in-plane.
ZnO nanowalls were excited from three different angles by rotating the sample in-plane.
Lasing from vertically-aligned ZnO nanowall

- ZnO nanowalls were excited from three different angles by rotating the sample in-plane.

![CCD image](image)

![Graph](graph)

Three angles of all shows different spectra.

The different oscillation routes are formed inside the nanowall.

Random lasing
Vertically-aligned ZnO nanowalls were synthesized by NAPLD method on ZnO buffer layer patterned by two-beam interference patterning.

The size of synthesized ZnO nanowall was that the height was about 7 µm, and the thickness was about 180 nm.

Lasing from a single piece of ZnO nanowall was achieved, and it had high crystallinity.

Lasing from the vertically-aligned ZnO nanowall was achieved for the first time, and the lasing mechanism is random lasing.