Nonlinear imaging of whispering gallery modes in GaN microwires

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Abstract. In this work non-scanning far-field nonlinear optical microscopy is employed to study the whispering gallery modes in tapered GaN microwire resonators. We demonstrate the confinement of whispering gallery modes under near-infrared excitation with the photon energy close to half of GaN bandgap. Our results indicate the enhancement of yellow-green luminescence by whispering gallery modes in GaN microwires.

1. Introduction

Epitaxially grown III-V nano- and microwires (MW) demonstrate smooth facets and high crystalline quality, and therefore, may be employed as naturally formed optical resonators for whispering gallery modes (WGM). Generally, WGM resonators are attractive structures for photonic applications due to their ultrahigh quality factors (Q). For example, recently, lasing in high-Q WGM cavities has been successfully demonstrated in hexagonal micro- and nanorods of GaN and ZnO [1,2].

Among III-V semiconductors, gallium nitride (GaN) is promising material for applications in nonlinear photonics in view of large second and third order nonlinear coefficients, wide direct bandgap (3.4 eV) and compatibility with high operational temperatures (melting point is 2500 °C) [3,4]. Large nonlinear coefficients measured in GaN [5,6] enable various nonlinear photonic applications such as signal processing [7], imaging and others [8].

Recently, the two-photon absorption (TPA) enhancement by WGM were observed [9] in GaN structures, which attracts additional interest to GaN NWs as TPA-compatible WGM cavities. Meanwhile, several studies of epitaxial bulk and thin-film GaN reported the enhancement of TPA around one half of the bandgap which allows efficient conversion from near-infrared (NIR) to ultraviolet visible range [5,6].

Spatial information on local field enhancement and structural defects is crucial for device design and operation diagnostics the MW-based applications [10,11]. This information may be accessed by far-field nonlinear optical microscopy, which is the fast non-invasive technique for high-resolution mode mapping in nano- and microresonators [12,13]. Here we use non-scanning far-field nonlinear optical microscopy to characterize the WGMs confined in epitaxially grown GaN MW resonators under NIR excitation.
2. Methods

2.1. MW synthesis
The samples of GaN MWs were synthesized by metal-organic chemical vapor deposition (MOCVD) on sapphire substrates covered by titanium films using a system with a horizontal flow reactor [14]. Ti films with a thickness around 14 nm were thermally deposited on the substrates prior to GaN growth. The MOCVD process started with 10 min heating in a nitrogen-ammonia atmosphere. After that, the atmosphere was changed to hydrogen-ammonia and trimethylgallium was introduced into the reactor chamber. The growth temperature was fixed to 1040 ºC, and the growth pressure was 200 mbar.

Morphology of the MWs was analyzed by scanning electron microscopy (SEM) in Magellan FEI 400 system. For optical characterization, the MWs were mechanically transferred on indium-tin-oxide substrates.

2.2. Optical characterization
Nonlinear optical studies of GaN MWs resonators were carried out in a home-built non-scanning transmission nonlinear optical microscope [13]. The MWs were placed in the focal plane of the incident polarized laser beam. The samples were excited with Ti:Sapphire tuneable laser source, that can operate in the region from 690 nm to 1000 nm. The laser was operated in pulsed regime with the pulse frequency of 80 MHz. The laser beam was focused with 10X objective and collected with 100X objective after passing thought the sample. Then the signal at excitation wavelength was filtered behind the sample with BG-39 filter (transmission range 300 – 700 nm). Output nonlinear signal was focused onto an CCD camera to get images. To measure the spectra from MWs excited with NIR laser source the setup was equipped with the SpectraPro imaging spectrometer. The intensity of the signal on each image was analyzed precisely in each pixel.

3. Results and discussion
In nonlinear imaging we collected the visible-range signal generated in MW resonator under NIR excitation at 740 nm. Panels (a) and (c-e) in Figure 3 show the SEM and CCD images of tapered GaN MW, respectively. Figure 1(f) characterize the MW tapering by diameter (D) dependence on the coordinate along the MW axis z.

CCD images in Figure (b-d) show the maximums of luminescence forms the three-dot patterns for specific positions along the MW axis. The intensive signal was also observed at the fractured ends of the MW. Images of the same MW in panels (b), (c) and (d) of Figure 1 correspond to three different positions of the excitation beam. By comparing CCD and SEM images we attribute the three dots of the luminescence patterns to the centers of MW facets and relate to WGMs confined at specific resonance diameters.

In tapered MW the resonant facet separations \( d_{WGM}^{m} \) for \( m^{th} \) WGM can be calculated as [15]:

\[
d_{WGM}^{m} = \frac{\lambda}{3n} \left( m + \frac{\beta}{\sqrt{3}n^2 - 4} \right),
\]

where \( \lambda \) is the wavelength, \( n \) is the refractive index and the parameter \( \beta = n \) for TM polarization and \( \beta = 1/n \) for TE polarization.

Figure 1(e) shows the resonance diameters \( D = 2d/\sqrt{3} \) for WGMs in MW resonator with hexagonal cross-section calculated using eq. (1). The calculated resonant diameters \( D_{WGM} \) match to three-dot intensity patterns in CCD images via the \( D(z) \) dependence. The \( D(z) \) was extracted from the fit of the diameters obtained from the analysis of the SEM image. We stress that matching can be obtained in the case of the second harmonic with \( \lambda = 370 \) nm.

Figure 1(f) shows the spectrum of the nonlinear signal collected from MW resonator. The spectrum has a broad yellow-green luminescence (YGL) band and the peak of second harmonic at 370 nm corresponding to double frequency of the excitation at 740 nm.

The YGL band in luminescence of GaN nanostructures is known to originate from the different types of the surface defect states [16,17].
Placing the 400 nm longpass FLG-400 filter between the sample and the CCD camera does change considerably the CCD images. Thus, the observed three-dot patterns can be related to YGL band. At the same time, mode matching shows that the three-dot pattern at the WGM resonance diameters. Therefore, we observe the enhancement of YGL by WGMs, as illustrated by schematic in Figure 1(g). These observations are consistent with recently reported increase of luminescence [18] and two-photon absorption [9] intensity related to confinement of WGMs.

Figure 1. SEM (a) and CCD images (b-d) and spectrum (f) of the MW resonator on indium-tin oxide substrate under 740 nm excitation. The CCD images are taken at three different beam positions (using only BG-39 filter). (e) Diameter of the tapered MW as the function of coordinate z along the MW axis and matching of WGM positions to resonance diameters.

Conclusions
In conclusion, we have shown that tapered GaN MWs, grown in Ti-assisted MOCVD process can be employed as WGM resonators. In this type of nanostructures, the confinement of WGMs was demonstrated under NIR excitation with the photon energy close to half of GaN bandgap. Far-field nonlinear imaging and spectroscopy were successfully employed as the tools for mapping and characterization of the confined WGMs. The obtained luminescence patterns witness the enhancement of YGL by WGM confined at resonance diameters of the GaN MW. We believe that our method of mode mapping will be useful for fundamental studies as well as for device diagnostics.

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