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Mapping of Fabry-Perot and Whispering Gallery Modes in GaN microwires by Nonlinear imaging

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Abstract

Engineering nonlinear optical responses at the microscale is a key topic in photonics for achieving efficient frequency conversion and light manipulation. Gallium nitride (GaN) is a promising semiconductor material for integrated nonlinear photonic structures. In this work, we use epitaxially grown GaN microwires as nonlinear optical whispering gallery and Fabry-Perot resonators. We demonstrate an effective generation of second-harmonic and polarization-dependent signals of whispering gallery and Fabry-Perot modes under near-infrared excitation. We show how the rotation of the excitation polarization can be used to control and switch between Fabry-Perot and whispering gallery modes in tapered GaN microwire resonators. We demonstrate the enhancement of two-photon luminescence in the yellow-green spectral range due to efficient coupling between whispering gallery, Fabry-Perot modes, and excitonic states in GaN. This luminescence enhancement allows us to conveniently visualize whispering gallery modes excited with a near-infrared source. Such microwire resonators can be used as compact microlasers or sensing elements in photonic sensors.

Keywords: GaN, second harmonic generation, mode imaging

1. Introduction

Semiconductor III-V nano- and microwires (MWs) are considered as building blocks for photonic and optoelectronic devices. Small footprint and effective strain relaxation on the sidewalls provide high crystalline quality in MWs and open up the opportunity for integration of wide-bandgap semiconductors with silicon platform [1–3]. GaN is one of the most promising III-V semiconductor materials for applications in nonlinear photonics due to strong second and third order nonlinear optical properties, direct bandgap (3.4 eV) and compatibility with high operational temperatures (melting point is 2500 °C) [4,5]. The non-centrosymmetric crystal structure of GaN exhibits prominent piezo-electric properties suitable for applications in integrated electro-optical components and sensing devices [6,7].

Epitaxially grown GaN MWs typically have a wurtzite crystal structure, a hexagonal cross-section and smooth side facets. Hence, GaN MWs naturally suit potential applications as optical resonators [8,9] for integrated lasers [10,11] and sensors [6,12]. Previous studies have shown how GaN nanostructures can be used for confinement of whispering gallery (WGM) and Fabry-Perot (FPM) modes [13–16].

Optical resonators supporting WGM can be applied for cavity devices with exceptional properties such as small mode volume, high power density, and narrow spectral linewidth [12,17]. Moreover, such cavity modes can be coupled to strong exciton resonances when their frequencies are close to each other [18–20]. However, the key challenge in the development of WGM resonators is to design and fabricate the nanostructures that will efficiently confine WGMs.

The earlier studies of FPMs in GaN and GaAs MWs have demonstrated efficient second-harmonic generation (SHG) [21–24], which can be applied to frequency conversion devices, including ultraviolet (UV) and visible range lasers integrated with near-infrared (NIR) sources. The III-V MWs are promising components for integrated nanosystems since they can be used as elements of both active devices and passive interconnectors [25]. One of the main advantages of GaN MWs is that they can be synthesized in scalable epitaxial processes on different substrates, including silicon. However, control and characterization of the modes excited in MWs resonators remain challenging and require new advanced approaches.

Previous works demonstrated nanoscale imaging using electron energy loss spectroscopy or cathodoluminescence [26]. Despite the high resolution of luminescence mapping, these methods rely on sophisticated electron microscopy equipment and use the excitation by the electron beam rather than optical pumping and thus cannot be used to study optical nonlinear effects. Recording of the nonlinear response at the nanoscale over a broad range of wavelengths and polarizations of the incoming excitation laser beam may require sophisticated scanning methods which are time-consuming and need expensive components [27].

Far-field nonlinear optical microscopy is a fast non-invasive non-scanning technique that allows high-resolution mode imaging in nano- and microresonators [28,29]. Spatial information obtained from mode mapping is essential for the studies of local field distribution and enhancement [30,31] as well as diagnostics of devices in operation [32,33].

Our study shows how epitaxially grown GaN MWs can be employed as WGM resonators in the nonlinear optical regime. We demonstrate how the far-field nonlinear imaging is used for non-invasive investigation and visualization of WGM and FPM coupled with excitonic states in GaN MW resonators under NIR excitation. Within this approach we also demonstrate how polarization of the pump excitation can be used to switch the intensity of FPM and WGM inside MW resonator. The aim of this study is to obtain new means of all-optical control and switching of the confined modes, which is in high demand for various nanophotonic applications, such as integrated lasers, photonic sensors, and optical circuits.

2. Results and discussion

2.1 Microwires synthesis and morphology studies

The GaN MW resonators were grown by metal-organic chemical vapor deposition (MOCVD) on sapphire substrates catalyzed by Ti films. The details of the fabrication method are presented in our previous work [34], and in Supplementary Materials (Section 1). The MWs are formed along the c-axis and show extraordinary high elongation rate, which reduces GaN deposition times down to 20 min [34] in comparison to few hours in standard MOCVD growth. The synthesized GaN MWs are slightly tapered and have symmetric hexagonal cross-section and flat top facets. Figure 1 (a) and (b) shows side and top-view SEM images of GaN MWs and figure 1(c) shows a MW mechanically transferred on an ITO grid.

Nonlinear optical measurements are typically sensitive to facet roughness [35]. Hence, the smoothness of MW sidewalls was verified by atomic force microscopy (AFM). The details of the AFM characterization are given in Supplementary Materials (Section 2).

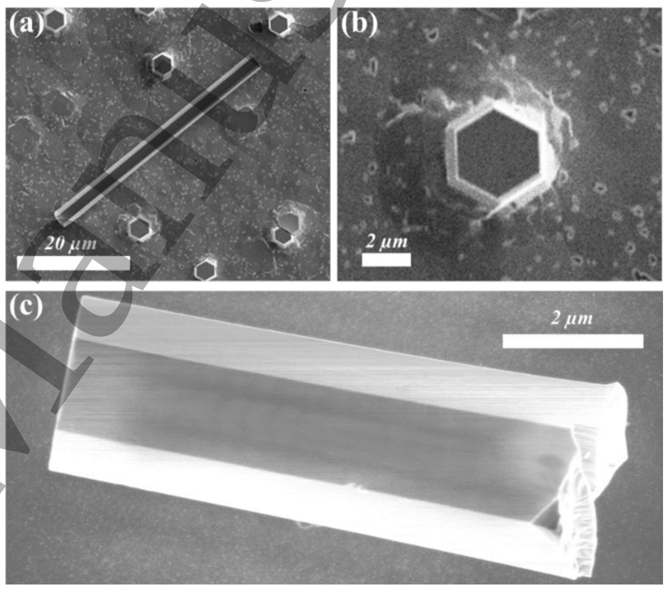

Figure 1. GaN MWs with hexagonal top facet on sapphire substrate (a) and (b). GaN MW after transfer to ITO grid for nonlinear optical characterization (c).2.2 Microwire optical characterization

A home-built non-scanning transmission nonlinear optical microscope [36,37] was used to characterize nonlinear optical performance of GaN MWs resonators. In the microscope the samples were excited with Ti: Sapphire pulsed (80 MHz) laser source that can operate in the region from 690 nm to 1000 nm. Additionally, the setup was equipped with a BBO crystal that allows to double the frequency of excitation from the NIR laser source to the UV range from 345 to 390 nm.

The samples of GaN MWs were transferred on the transparent ITO substrates and placed in the focal plane of the incident polarized laser beam. The setup allows one to vary the polarization of the incident beam from parallel to the long axis of the MW to perpendicular to it. In the output signal, we acquired the spectra using the spectrometer and the spatial mode images with an electron-multiplying charge-coupled device (CCD) camera. Thus, we investigated linear and nonlinear optical responses of the synthesized MWs from UV and NIR excitation, respectively. In this study the linear optical response was obtained under UV excitation at 370 nm, which is close to the wavelength equivalent of the GaN bandgap [5].

The nonlinear response from GaN MWs was obtained under NIR excitation of 740 nm, about twice the wavelength of the bandgap. Thus, the recorded nonlinear response was stimulated by the absorption of two photons. After the sample, the NIR excitation wavelength was filtered out from the output signal. More technical details of sample preparation and optical measurements are given in Supplementary Materials (Section 1).

Figure 2 (a) and (b) shows the overlaid SEM and CCD images obtained under UV and NIR excitation, respectively. In both cases the fractured regions at the top and the bottom of the MW show more intense luminescence than the central parts of the MWs. Under UV excitation, the central parts of the MWs show the bright lines at the centers of three MW facets (figure 2(b)). Meanwhile, under the NIR excitation, the maxima of intensities are observed only at the specific diameters within the central parts of MWs (figure 2(a)). The signal in figure 2(a) presents the compact pointed three dots patterns that are visible in the centers of MW facets that we relate to the WGM confined in the MW resonator.

Figure 2 (c) shows typical examples of spectra of the tapered GaN MW acquired under UV (370 nm) and NIR (740 nm) excitation. The corresponding spectra in figure 2(c) demonstrate broad yellow-green luminescence (YGL). In previous studies, the YGL was attributed to defect states concentrated at the surface of GaN MW sidewalls [38,39]. Besides the YGL band, the spectrum under NIR excitation shows the peak around 370 nm, which combines the SHG at half of the incident field wavelength and the near band edge (NBE) emission.

The spectra and CCD images under NIR excitation are sensitive to the polarization of the incident laser beam, as shown in figure 3 (a, b). The mode imaging in figure 3(a) shows the shape variation of the luminescence patterns with the change of excitation polarization. The three-dot patterns have the maximum intensity when the incident light is polarized along the MW axis (angle $\theta = 0°$) and are not visible when polarization of the incident beam is 90° rotated.

The spectra in figure 3(b) were obtained after filtering most of the incident NIR light and normalized by the intensity of unfiltered incident field (740 nm). The peak near 370 nm corresponding to SHG and NBE signal shows only slight intensity variation with the change of excitation polarization. Meantime, the intensity of the YGL peak varies from almost zero to twice the intensity of the unfiltered incident field.

Figure 3(c) shows that the normalized intensities of the three-dot pattern in CCD images (blue circles labelled "CCD image") are well-correlated with the normalized intensities of YGL parts of spectra (red squares labelled "Spectrum") as functions of polarization of the incident field. The grey solid line in figure 3(c) shows the cosine-squared function which gives a good approximation for both dependencies. In contrast to NIR excitation, both spectra and CCD images show no dependence on the polarization under UV excitation at 360 – 370 nm, which is illustrated in figure S3 in Supplementary Materials (Section 3).

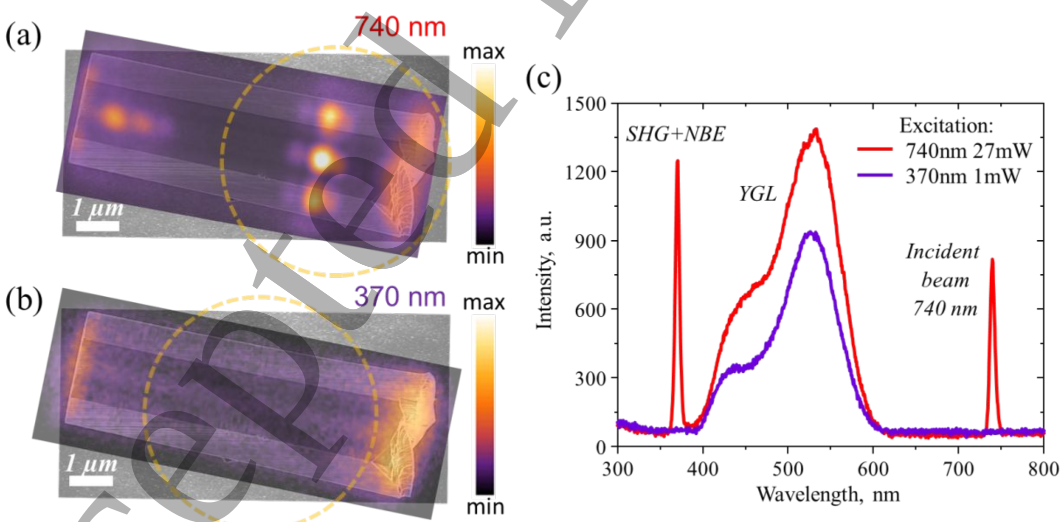

Figure 2. SEM image of GaN MW matched to CCD images acquired under NIR (a) and UV (b) excitation. The dotted circles show the approximated width of the laser beam. (c) photoluminescence spectra of GaN MW under UV excitation at 370 nm and NIR excitation at 740 nm.

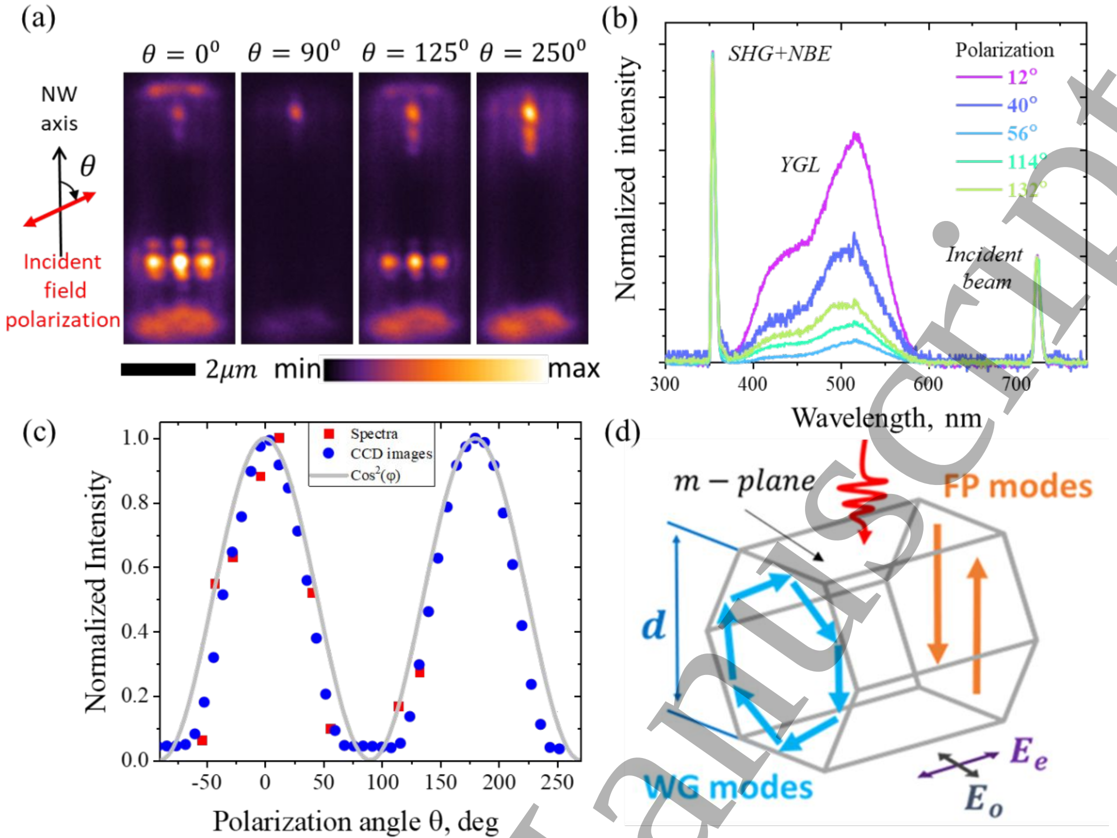

Figure 3. CCD images (a) and spectra (b) obtained under NIR excitation (740 nm). (c) Normalized intensities of YGL peaks in PL spectra (red squares labeled "Spectra"), the normalized intensity of three-dot pattern in the nonlinear CCD images (blue circles labelled "CCD image") at different angles of NIR (740 nm) excitation polarization. Error bars are comparable with the symbol size. (d) Schematic of WG and FP modes in wurtzite GaN MW resonator.

Next, we analysed the excited modes in GaN MW resonators to match the observed intensity patterns to the corresponding FPMs and WGMs. The confinement of FPMs and WGMs was earlier demonstrated in various GaN nanostructures with hexagonal cross-section: nanowires, nanorods, nanopyramids, etc. [13,14,17,40]. Figure 3(d) illustrates schematically the FPMs and WGMs in wurtzite GaN MW resonator. The size of the MW resonator is characterized by the distance between $\{10\bar{1}0\}$ facets (m-planes) denoted as d. In tapered MWs with d continuously changing along the wire axis, FPMs and WGMs contribute to the confinement of hybrid modes [41,42]. The modes of each of two types are characterized by the resonant facet separations d_{FP}^m and d_{WG}^m for m^{th} FPM and WGM, respectively. Within the plane-wave model the resonant diameters of GaN MWs can be calculated as [43]:

$$d_{FP}^m = m\,\lambda/2n \qquad (1)$$

$$d_{WG}^m = \frac{\lambda}{3n}\left(m + \frac{6}{\pi}\left(\beta\sqrt{3n^2-4}\right)\right), \qquad (2)$$

where λ is the wavelength, n is the refractive index and the parameter $\beta = n$ for TM polarization and $\beta = 1/n$ for TE polarization. We study wurtzite GaN MW growing along the c-axis associated to the extraordinary direction with $n = n_{ext}$ (TM). Thus, the ordinary direction with $n = n_{ord}$ (TE) is perpendicular to the MW axis as shown in figure 4(a).

The observed nonlinear YGL and UV luminescence under NIR excitation requires absorption of two or more photons simultaneously. Previous studies report considerable excitonic enhancement of two and three-photon absorption in GaN [44–46]. Therefore, the most effective FPM and WGM excitation is expected when coupled with excitonic state.

Previously, strong coupling between the exciton states and photonic cavity modes in GaN and ZnO was observed from cryogenic up to room temperatures [47–49]. From a quantum mechanical point of view the mixed state can be characterized by the superposition of excitonic and photonic wave functions [50]. Thus, the interaction between the cavity photons and excitons can be described in terms of exciton-polariton quasiparticles with energy E_p. Strong coupling in GaN microcavities results in splitting to two modes called upper

(UP) and lower polaritons (LP) with the energies about 3.40 eV and 3.35 eV, respectively [19]. Figure 4(a) shows the schematic diagram for transitions associated with polaritonic states [51].

Thus, we estimate the resonant facet separations for FPMs and WGMs (plotted in figure 4(c)) by inserting $\lambda = E_p/hc$ with E_p for UP or LP into eq. (1-2). Then we match the calculated facet separations with corresponding positions along the MW axis according to observed MW geometry. Circles in figure 4(d) show the measured facet separations d and their position along the MW axis extracted from SEM image. The polynomial fit was used to estimate the positions of FPMs and WGMs positions in the tapered GaN MW. These estimated mode positions match to the intensity maximums (dots patterning) in the CCD images in Fig 4(b). Thus, we can map the three-dot patterns to WGMs and single-dot maximums to FPMs.

Figure 4(b) shows that the observed features of the nonlinear response of GaN MWs are not specific for 740 nm excitation and were observed under the excitation with shorter and longer wavelengths. Figure 4(b) shows two clearly distinguishable three-dot patterns of WGMs coupled with excitonic states. More examples of spectra and CCD images for excitation at 770 nm are given in figure S4 in Supplementary Materials (Section 4).

The CCD images in figure 3(a) and figure 4(b) show only one or two WGMs which corresponds to the position of the exciting laser beam spot next to the wide end of the tapered MW. The other WGMs could also be visible when moving the position of the laser beam spot within the MW length, as illustrated in figure S5 (Supplementary Materials, Section 5).

The intensity patterns matched in our analysis to FPMs and WGMs depend differently on polarization of the incident light. Figure 3(a) shows three types of CCD images acquired at different excitation polarization: (i) when only FP mode is excited ($\theta = 90°$); (ii) when only WG mode is excited ($\theta = 0°$) or (iii) when both types of modes are excited ($\theta = 125°$). Thus, in the tapered GaN MWs studied in our work, the variation of polarization of NIR excitation allows selective excitation of FPM, WGM or both modes simultaneously. This approach gives the means for an all-optical switch of the resonance modes in hexagonal GaN resonators. Previously the selective excitation and switching between FP and WG modes were demonstrated by Baek et al. [52] where the selectivity was achieved by shifting the position of excitation from the center of the wire to its edge. Furthermore, the approach of [52] is limited to wires smaller than 1.5 μm in diameter and demonstrated only for UV excitation. Our approach uses NIR excitation and is suitable for large rods while the tapered shape allows switching without changing the position of excitation by varying the incoming polarization.

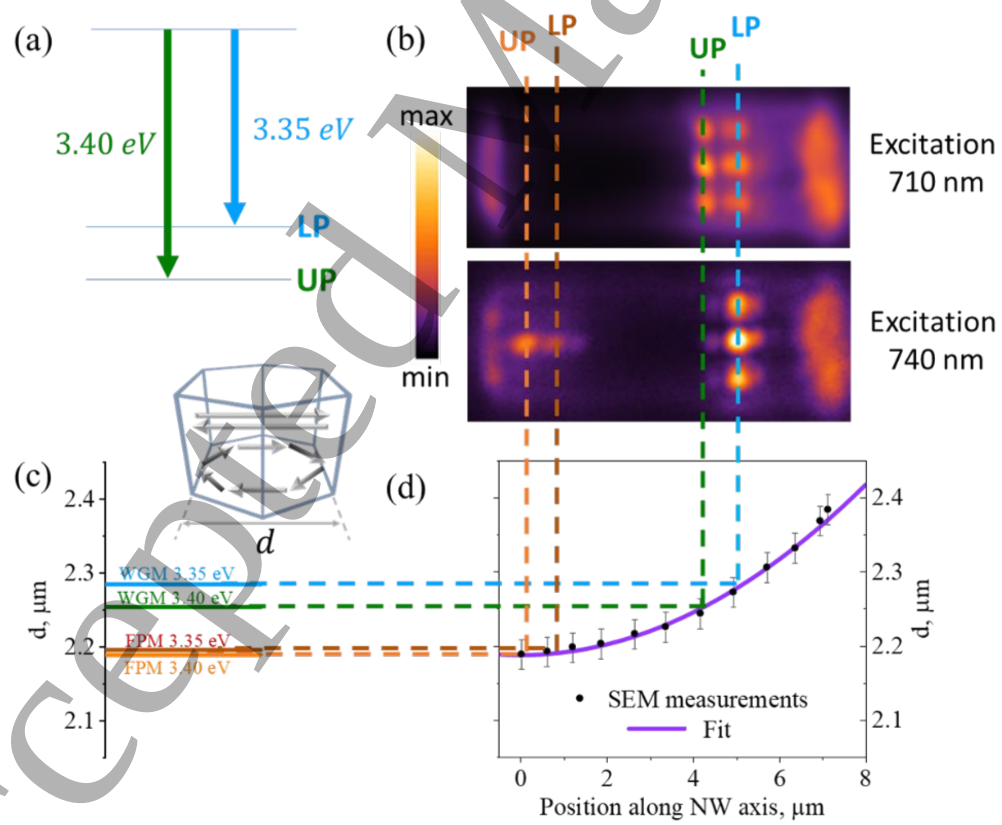

Figure 4. Schematic diagram for polaritonic transitions (a). Resonance facet separations (c) matched to the dotted patterns in CCD images (b) via the dependence of facet separation on position along the axis of the tapered MW(d).

We have found that the spectra of dotted patterns include the YGL band. As we show in Supplementary Materials (Section 6) the dotted patterns of luminescence maxima remained visible when the 400 nm long-pass filter was placed between the sample and the CCD camera. As we have shown in figure 4, the position of the three-dot pattern intensities corresponds to the WGM resonance diameter. Thus, we conclude that the coupling between excitonic states and WGMs excited under NIR irradiation results in local enhancement of the YGL at resonant diameters. In our interpretation we assume Purcell enhancement of the defect-related yellow-green luminescence. Each defect in GaN is introduced either by Ga-vacancies or C incorporation at the surface of the microwire sidewalls is considered as a compact light source. C-related defects in GaN nanostructures grown via a MOCVD process are known to originate from trimethylgallium precursor molecules [38]. Emission strength of these light sources is considered to be proportional to the photonic density of states induced by the cavity modes.

On one hand, the presence of YGL is widely considered as an undesirable factor related to defect states [1,53,54]. Except for some attempts of controlled use of artificially created defects [55,56], most studies of YGL aim at the quenching of defect luminosity [39,53]. On the other hand, in this work for the first time the YGL serves as a tool for visualization of WGMs coupled with excitonic states in the nanostructures, which cannot be observed directly. In hexagonal GaN MW cavities the WGM intensity has maximums in the directions along the MW facets [57], and minimums at the 90° angle to the m-plane facet (see figure 4(c)). In our setup we collect the signal in the latter direction, meanwhile, the YGL enhanced by WGM allows its visualization at the normal orientation to the sidewall plane.

3. Conclusions

In conclusion, we demonstrated that the Ti-assisted growth of GaN MWs by MOCVD provide the means for fast fabrication of optical FPM and WGM microresonators. We presented the approach to visualize WGMs and FPMs in tapered GaN MWs. Optical modes can be mapped and characterized via far-field nonlinear imaging and spectroscopy under UV and NIR excitation. We imaged WGMs and FPMs coupled with excitonic states excited with NIR sources. The obtained CCD images show that the patterns of nonlinear luminosity correspond to YGL enhanced at WG and FP resonant diameters. We demonstrated the selection between FPM and WGM confined in the tapered GaN MWs controlled by polarization of the incident laser beam. We believe that the suggested method of nonlinear optical characterization may be implemented for further studies of the light-matter interaction in semiconductor MWs. GaN MWs characterized in our work can be used as all-optical switching resonators supporting the high-quality WGM modes. Such MW resonators can be applied as a building blocks for compact frequency converting WGM microlasers, optical sensors and other light sources that can be controlled by the excitation polarization.

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