Characterization of optical and nonlinear properties of individual GaP nanowires using optical tweezers

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Abstract. Semiconductor nanowires (NWs) offer multiple advantages for designing novel optoelectronic devices, such as small footprint, high quantum efficiency, high nonlinear susceptibility. Gallium phosphide (GaP) is one of the attractive materials owing to its low optical absorption and high nonlinear susceptibility. However NWs should be transferred to planar substrates for optical studies, which do not allow efficient signal outcoupling. We demonstrate efficient second harmonic generation in individual GaP nanowires trapped using optical tweezers. Such vertically arranged configuration of NW allows to both efficiently generate second harmonic and to probe linear optical response using broadband light source. Such experiment allows to examine interplay between harmonic generation efficiency and NW dimensions.
1. Introduction

Demands for miniaturization of photonic devices promotes using single nanostructures as functional elements of the circuits. Semiconductor nanowires (NWs), falling between zero-dimensional (such as quantum dots) and two-dimensional structures, offer superior charge confinement while still remain electrically and optically accessible. NWs have been demonstrated as a promising platform for single-photon emitters [1], detectors and converters [2] and many others.

III-V semiconductors offer distinctive advantages, including bandgap engineering[3] and tailored electronic properties[4], which are even further enhanced by reduced dimensionality of NWs. III-V NWs are commonly synthesized using epitaxial techniques either on native or on silicone substrates [5]. Characterization of as-grown NWs is limited owing to high refractive index of the substrate, which makes light leakages to the substrate pronounced [6]. Owing to their small footprint, NWs can be easily mechanically cleaved and transferred to substrate with lower refractive index, like glass. Such planarization allows for analysis of single-NW photoluminescence, Raman and dark-field scattering. However such planar arrangement does not allow efficient excitation along the NW growth axis, which would allow studies of NW nonlinear response and waveguiding properties. Encapsulation in optically transparent membrane with subsequent detachment is one of the approaches towards efficient excitation of NWs along axis [7]. However, owing to small NW cross-section and their high density, spectroscopic studies of individual nanowires becomes complicated.

Optical tweezers[8], initially developed for manipulation of spherical objects, has been demonstrated as a tool for manipulation of nanostructures with nontrivial shapes. Optomechanical manipulation of NWs has been demonstrated in several reports primarily aimed for studies of fluid interactions [9]. Efficient second harmonic generation was demonstrated in individual potassium niobate NWs [10] and microphotoluminescence response was studied for individual InP NWs [11].

However, some important aspects were overlooked, such as impact of NW geometry on the nonlinear generation efficiency. Simultaneous probing of individual optically trapped GaP NWs with several laser sources allows to implement simultaneous study of linear and nonlinear response. We experimentally explore correlations between NWs geometry and its SHG response.

2. Results and discussion

GaP NWs were synthesized using Veeco GEN III molecular beam epitaxy machine on Si(111) substrates according to protocol described elsewhere [12]. Typical NW sizes are 10\(\mu\)m length and 175nm diameter. For the optical studies in the optical tweezers NWs were mechanically cleaved from substrate using ultrasonic bath and were transferred in the aqueous solution. Obtained solution contains NWs of different lengths caused by presence of shorter NWs on the growth substrate and uncontrolled breakdown during sonicaton.

The fluid cell with NWs solution was placed in optical tweezers setup with enhanced spectroscopic capabilities. The setup uses high-NA oil immersion objective for simultaneous trapping and probing of NWs. Setup schematics is depicted in Fig. 1(a). 980nm CW laser diode (Thorlabs) was used for NW trapping. Nonlinear response was measured using femtosecond 1030nm laser (YLMO 2W, Menlo Systems). Linear optical response was probed using spectrally filtered supercontinuum (YSL SC-Pro with VLF tunable filter, YSL Photonics) with filter band set to 550-700nm to avoid spectral overlap with SHG signal around 515 nm. Backscattered light was collected and analyzed using fiber-coupled spectrometer (Avantes).

The image of free-floating NW is shown in Fig.1(b). The length of the individual NW can be assessed both from optical image and from spectroscopic studies. Typical image of observed SHG pattern from single NW is shown in Fig.1(c). The integral intensity of SHG image can be used for further comparison of SHG efficiency between individual NWs.

Acquired spectrum demonstrates Fabry-Perot (FP) resonant modulation, confirming, that the NW acts as an optical cavity (Fig.2(a)). Lorentzian fit of individual dips in the spectrum allows to extract Q-factors of the resonance, which range between 150 to 200. Such values are reasonable for weak
lateral mode confinement. It is worth noting, that generated SHG signal can be modulated by guided modes of NW as well.

FP modulation of the scattered signal allows to assess the length of NWs, considering

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

where \( L \) is NW length, \( \lambda \) – wavenlength, \( \Delta \lambda \) – mode spacing, \( n \) – NW refractive index.

![Experimental setup schematics](image)

**Figure 1.** (a) Experimental setup schematics. (b) Microphotograph of free-floating single NW. (c) Second harmonic emission pattern of optically trapped NW

Several NWs were imaged optically and characterized with FP formula, showing good correlation between the data from the optical images and spectroscopic data.

The information on SHG intensity was obtained by integrating the intensity values of obtained SHG images of individual NWs. The summary on SHG intensity on NW length is shown in Fig 2(b). No clear dependency between NW length and SHG signal is seen. However, diameter of NWs plays crucial role in SHG efficiency. The dispersion of the diameters of NWs falls in range of 150 to 250 nm. However, an accurate numerical modelling is required to reveal interplay between SHG efficiency and NW diameters.
3. Conclusion

In summary, MBE-grown GaP nanowires grown on Si(111) were spectroscopically studied. The growth protocol allowed to synthesize the NWs with a broad distribution of the dimensions of individual NWs. Systematic study of linear and nonlinear properties of individual NWs can be performed by simultaneously probing them with broadband 550-700nm supercontinuum source and 1030nm femtosecond laser pulses. The modulation of linear spectroscopic response allows to obtain information on the longitudinal dimensions of the NW acting as FP cavity and to excite SHG and compare nonlinear response for different individual NWs. The obtained data demonstrates no direct correlation between NW length and SHG efficiency. NW diameter should be considered as an important parameter for accurate characterization of SHG efficiency.

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