Silicon nanowire/polymer membrane for infrared visualization via third-harmonic generation

Anna Nikolaeva,1, a) Vladimir V. Fedorov,2 Alexander S. Gudovskikh,2 Ivan A. Morozov,2 Viktoria Mastalieva,2 Vladimir Neplokh,2 Ivan S. Mukhin,2 and Sergey V. Makarov1

1)Department of Physics ITMO University St. Petersburg 197101 Russian Federation.
2)Aferov University St. Petersburg 194021 Russian Federation.

a)Corresponding author: anna.nikolaeva@metalab.ifmo.ru

Abstract. Nonlinear harmonic generation in nanostructures is one of the key topics in nanophotonics, as it allows infrared-to-visible light conversion at the nanoscale. This work reports on the efficient third-harmonic generation in a free-standing Si nanowire array encapsulated into a polymer membrane. High nonlinearity of Si material $\chi^{(3)}_{1111} \approx 2.45 \cdot 10^{-19} \text{m}^2/\text{V}^2$ and light coupling with optical resonances in the nanowires yield a strong third-harmonic signal and efficient infrared (1200 – 2000nm) to visible (400 – 660nm) upconversion. The fabricated membranes demonstrate high flexibility and semitransparency, which makes them convenient to use as visualizers.

INTRODUCTION

Observation and use of nonlinear optical effects became possible after the advent of powerful laser sources, but the effects themselves are rather weak, which requires new approaches for the enhancement of their generation. Recently, dielectric nanostructures with intrinsic nonlinearity appeared to be prospective objects for signal enhancement [1], where third-harmonic generation (THG) efficiency can reach values of $10^{-7} – 10^{-5}$ [2, 3] in a silicon-based structure. Third-harmonic generation from resonant nanostructures has become a powerful tool for nanoscale optical characterization [4], shaping of the directionality of nonlinear radiation [5], non-linear holography [6] as well as for the creation of nanoscale color-tunable light sources via infrared (IR)-to-visible light conversion [7].

In this work, we took advantage of the work [8], namely, that the increase of the THG conversion efficiency can be achieved with elongation of the nanoparticles in one direction, forming nanowires [9]. In such designs, a combination of Mie resonances and Fabry-Perot modes can lead to strong incident field localization and outstanding optical properties. But we use silicon material and the THG process to visualize longer wavelengths.

EXPERIMENTAL DETAILS

Sample fabrication. The optical properties have been studied in arrays of vertically oriented SiNWs encapsulated into a silicone membrane and peeled from the Si substrate. This technology enables the reuse of growth substrates and a combination of various material systems to create ultra-thin (up to several microns thickness) flexible transparent structures. “Black silicon” NWs were produced by the cryogenic process. Plasma etching of Si (100) substrates under ~140°C temperature is used and the NW side surface is passivated due to the formation of a non-volatile SiOxFy compound at cryogenic temperature. It blocks the etching mechanism and the cryogenic process provides deep and anisotropic etching of silicon [10]. The height of obtained NWs for $X_1$ sample was about $h = 9 \mu m$, while diameter was in the range of $d = 300 – 1350 nm$ for $X_2$ sample, what is shown in the scanning electron microscopy (SEM) images in Figure 1(a,b). Further, the NW arrays were encapsulated into a Polydimethylsiloxane (PDMS) matrix and separated from the growth substrate.

Nonlinear Optical Characterization. We perform nonlinear optical characterization of the fabricated membranes in transmission geometry with a custom nonlinear optical setup. A detailed schematic of the experiment is presented in Figure 1(c). Tunable sample excitation in the range of 1200-2000nm was performed by a femtosecond Pharus laser and Optical Parametric Amplifier (OPA) ORPHEUS-HP. In our experiments, the membranes were placed on glass coverslips and aligned perpendicular to the laser propagation axis. The laser light was focused with a 50x objective (NA = 0.42) on the sample. The signal was then collected with a 50x objective (NA = 0.55) and focused onto a scientific CMOS camera. The power was controlled via a half-wave plate placed on the excitation beam path before...
FIGURE 1. (a,b) SEM images of studied Si NW: sample $X_1$ highlighted in blue frame (a) and sample $X_2$ highlighted in red frame(b). c) Schematic of experimental setup for third-harmonic generation spectroscopy.

the Glan prism as shown in Figure 1(c). For the nonlinear optical characterization, we performed the measurements of THG intensity for different excitation wavelengths at same average power $P_{av} = 3mW$ (Figure 2(b,e)) and for different average pump power at the same excitation wavelength $\lambda = 1400nm$ (Figure 2(a,d)).

RESULTS AND DISCUSSION

To demonstrate efficient infrared-to-visible light visualizers based on SiNW/polymer membranes having THG signal, we studied THG from the SiNWs. First, we measured the intensity of the signal from the fluence, which we showed in Figure 2(a,d) on a double logarithmic scale. After the fitting, we got a slope of approximately 3, which ensures that we have measured the third-harmonic generation. The bend in Figure 2(d) is probably because the PDMS was warming up and the structure went out of focus, which was visible on the CMOS camera. Our structure works as an efficient visualizer over a wide range of pump wavelengths 1200 $\text{−} 2000$nm, it is shown in Figure 2(b,e), which shows the dependence of the THG intensity on the pump wavelength, and in Figure 2(c), showing the CMOS camera images of THG signal for different pump wavelengths.

Remarkably, optical resonances (Fabry-Perot and Mie-type) in SiNWs support strong incident light localization at certain wavelengths, resulting in the appearance of pronounced maxima in Figures 2(b,e). This effect helps to enhance THG and it is generally an advantage of resonant nanostructures, allowing for the creation of efficient and thin optical devices. Moreover, our SiNWs support the resonances in a broad spectral range due to the dispersion of the diameters and heights of nanowires, also, nanowires with varying morphologies interact differently with pump light and thus exhibit a broad spectrum of third-harmonic generation. In consequence, the conversion efficiency is homogeneously distributed over near-IR.

CONCLUSION

In this work, we have demonstrated efficient infrared-to-visible light visualizers based on SiNW/polymer membranes having THG signal. For this purpose, we have employed the strong THG process in the resonant SiNWs in a broad spectral range. Such SiNW structures enabled the development of thin 10–20$\mu$m membranes with efficiently converting IR radiation into visible light. The proposed THG-based principle of IR visualization differs in both functional and operational characteristics from most commercial analogs made of a special ceramic with anti-Stokes luminophores.
FIGURE 2. Experimental characterization of THG from SiNW/PDMS, (a,b,c) corresponds to $X_1$ sample and (d,e) to $X_2$. (a,d) Experimental dependence of the THG intensity on fluence at a wavelength $\lambda = 1400$nm and repetition rate of $RR = 1$MHz (stars); blue and orange line is the result of the fitting procedure. (b,e) Experimental dependence of the THG intensity on the pump wavelength. Up to 1500nm, we used a FESH750 short-pass filter and then we changed it to FESH1000. c) $X_1$ sample image captured with the CMOS camera in the light of the white lamp, as well as images of third-harmonic generation signals for different pump wavelengths $\lambda = 1440$, 1480, 1520, 1600, 1780, 1840nm.

In contrast to the anti-Stokes luminescence frequency conversion, the THG process is less temperature-dependent and makes it possible to distinguish the IR pump wavelength by the color of its second harmonic. Also, as compared to the ceramic-based IR viewers suffering from hygroscopicity problems, the SiNW array can be easily fully encapsulated in silicone elastomer, protecting them from the environment. Weak absorption of the pump radiation together with high thermal conductivity of the Si material makes the fabricated membranes suitable for high power density applications.

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