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Broadband photoluminescence of silicon nanowires excited by near-infrared continuous wave lasers

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In advanced nanomaterials field, silicon nanowires (SiNWs) play an increasing significant role due to the outstanding optical properties. Although various kinds of investigations for SiNWs in optical characteristic have been proposed, it remained rare study of the photoluminescence (PL) phenomenon. Here, we theoretically and experimentally demonstrate an upconversion PL with broadband spectrum by exciting SiNWs using 980 nm continuous wave laser. PL spectra are efficiently detected and range from 500 nm to 920 nm. An electron can be transferred to higher excitation energy levels by absorbing one photon with the assistance of multi-phonons, producing hot luminescence. The proposed concept of PL phenomenon can be extended to biosensor, fluorescence labeling systems, and miniature broadband optical source emitters.

1. Introduction

Silicon nanowires (SiNWs) have attracted enormous attention due to their potential applications in optical [1], nanoelectronic [2] and thermoelectric fields [3]. For now, the investigation of SiNWs not only contribute to the availably nanostructures [4] and optical properties [5–7], but also the significantly strong room-temperature photoluminescence (PL) and quantum size effects. Actually, based on the great potentials in SiNWs thin-film solar cells, the PL phenomenon and mechanisms of SiNWs have aroused extensive attentions. Initially, an unstable bright red PL was first observed for porous Si [8] and shifted into the visible for crystallite sizes below 5 nm while the quantum size effects theory was probably proposed to account for the mechanisms [9]. Because of the indirect band gap of Si, the weak light absorption and radiation in the visible light range have interdicted the further applications in photonics and so on [10]. Bandiera et al. [6] demonstrated that, with the periodic structures properly designed, the nanostructured Si could enhance the light absorption. The experimental result exposed that the crystalline-amorphous core-shell silicon nanowires exhibited extremely high absorption of 95% at short wavelengths ($\lambda < 550$ nm) and a concomitant very low absorption down to less than 2% at long wavelengths ($\lambda > 780$ nm) [7]. Excited by 337 nm and 488 nm laser light, Sivakov et al. reported a strong visible red-orange PL of rough silicon nanowire sidewalls at room temperature [4]. Ledoux et al. explored the mechanisms of PL based on different-size-SiNWs and presented a theoretically model to describe the quantum confinement of size to the PL for SiNWs [11]. Jing et al. also studied the effects of geometry on the mechanical properties of SiNWs and found that the fracture stress decreased as the periodic length increased [12]. These useful findings can provide insights into SiNWs fabrication and development of their applications.

Very recently, the results proved that, through hot-electron-assisted photocatalysis, the plasmonic nanoparticles could enhance the significant promise for solar energy [13]. Interestingly, the hot-electron-assisted catalysis efficiency was correlated with the intensity of the photoluminescence (PL) of gold nanoparticles and silver films [14]. It was found that such hot spots could emitted efficiently both up- and down-converted hot luminescence under excitation of femtosecond laser pulses. Different with the

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other upconversion physical mechanisms, the luminescence from such hot spots originated from the black-body emission of hot electrons generated by intraband transitions [15]. For SiNWs, the hot luminescence was confirmed to originate from the intraband transitions of electrons by using excitation photon energy smaller than its band-gap energy. To the best of our knowledge, the upconversion emission of SiNWs excited by the near-infrared light has not been investigated.

In this work, we have presented a broadband three-peaks-structure upconversion PL spectra of SiNWs excited by 980 nm continuous wave (CW) lasers. The upconversion physical mechanisms were also proposed to account for the main feature of the experimental phenomenon. The morphological characteristics of SiNWs were highly performed utilizing the SEM and Raman spectrometer. The spectra characteristic demonstrated that SiNWs could be rationally designed as light source emitters, which has significantly potential application for broadband optical source, biosensor, fluorescent labeling systems and so on.

2. Experimental details and discussions

The SiNWs were synthesized by metal-catalyzed electroless etching (MCEE) [16]. The detail fabrication was elaborated as follow: the n-Si (100) wafer pieces were cleaned by ultrasonically rinsing in acetone for 5 min and followed by acetone rinse for another 5 min. After the Si wafer pieces were boiled in H_2SO_4:H_2O_2 (3:1) for 30 min, they were guaranteed to be cleaned by rinsing with adequate deionized water (DW). The aqueous solution, 0.02 moles of silver nitrate ($AgNO_3$) and 5 moles of hydrofluoric acid (HF) in the volume ratio 1:1 solution, in sealed container was utilized to steep the cleaned Si wafer. The SiNWs were produced vertically after 30–60 min. Another copy of H_2SO_4-H_2O_2 solution was contributed to eliminate the silver dendritic on SiNWs surface. Finally, the surfaces obtained after the etching procedures were rinsed several times in DW and dried at room temperature. Structures analysis of SiNWs has been carried out by a JSM-7001F scanning electron microscope (SEM). Fig. 1a and b showed the as-prepared SiNWs array with the uniform length of ~6 μm and diameter in the range of 30–100 nm. The front view of SiNWs presented that our fabricated samples distributed uniformly in large area and cluster partly in small region. Fig. 1c gives out the UV–visible absorption spectra by using a PerkinElmer Lambda 950 UV–Vis-NIR spectrophotometer. From Fig. 1c, it can be seen that the absorption spectra of SiNWs are quite flat in the visible light range with a smooth peak appearing at about 400 nm. The results show that the SiNWs have high absorption efficiency in all visible regions. Scraped off from the silicon substrate in the alcohol solution, Fig. 2d exhibits the Raman spectra of the SiNWs samples under excitation of the CW laser with central wavelengths of 633 nm. The spectra shows remarkable Raman features at 519.9 cm^{-1}, which is good agreement with the theoretical calculations and experiments [17]. The Raman peak also shows a high purity SiNW samples without other impurities.

The PL experiments were carried out using a 980 nm laser for excitation at room temperature. Fig. 2 depicts the schematic illustration of the experiment. In order to eliminate the negative effect of the silicon substrate, the SiNWs were scraped off from the silicon substrate and mixed into absolute ethyl alcohol (EtOH). The finally SiNWs-EtOH suspension solutions with different concentration were obtained for PL. Then, an appropriate suspension was drawn onto a clean glass slide with a pipette. A CW laser light

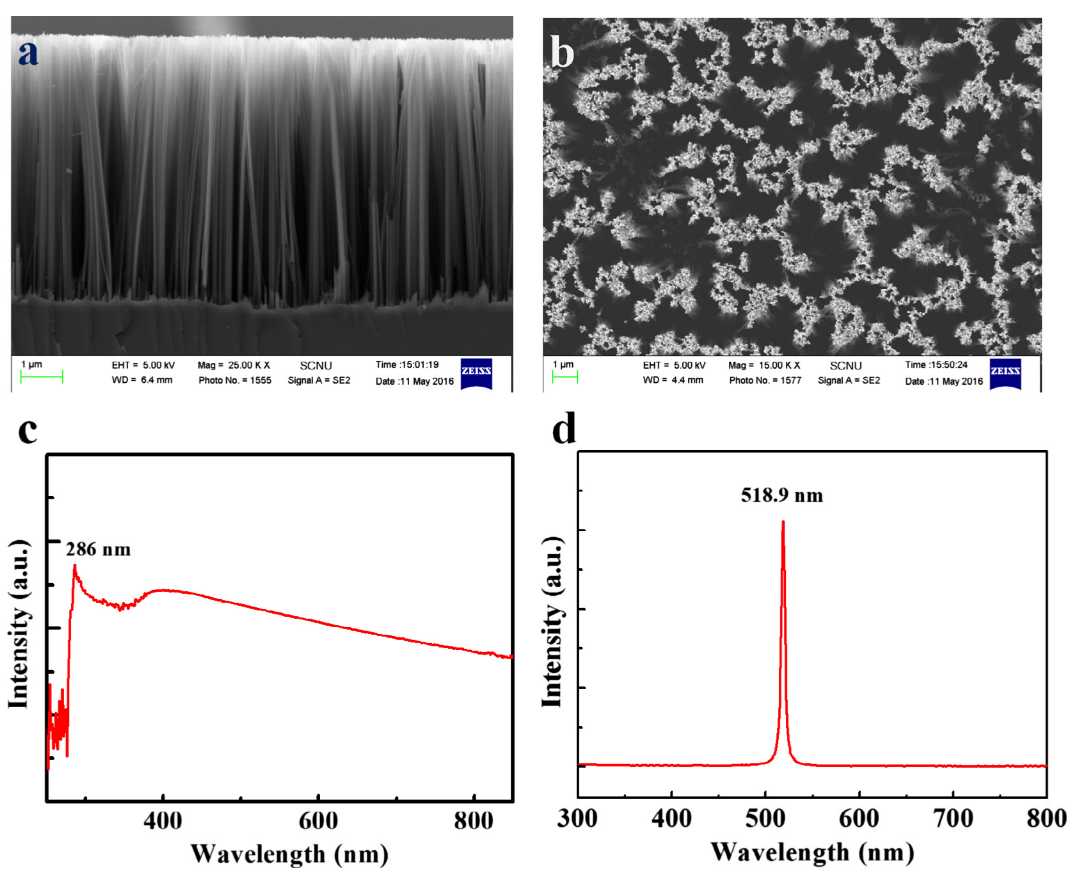

Fig. 1. Characteries of SiNWs. Typical SEM cross-section (a) and front-view (b) SEM images of SiNWs array after 5 cycles of DR process. (c) UV–vis spectra of SiNWs. (d) Raman spectra of SiNWs excited by 633 nm laser light source.

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