Optical gain based on NaYF₄: Er³⁺, Yb³⁺ nanoparticles-doped polymer waveguide under convenient LED pumping

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^(b) Zhaoqin Zhou, Jiabi Xue, ^(b) Baoping Zhang, Ce Wang, ^(b) Xingchen Yang, Wang Fan, Leiying Ying, ^(b) Zhiwei Zheng, Yongjun Xie, Yuanfei Wu, Xiaodong Yang, ^(b) Dan Zhang, et al.



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Zhaoqin Zhou,¹ D Jiabi Xue,¹ Baoping Zhang,¹ D Ce Wang,¹ Xingchen Yang,¹ D Wang Fan,¹ Leiying Ying,¹ Zhiwei Zheng,¹ D Yongjun Xie,¹ Yuanfei Wu,² Xiaodong Yang,² and Dan Zhang^{1,a)} D

AFFILIATIONS

¹School of Electronic Science and Engineering (National Model Microelectronics College), Xiamen University, Xiamen 361005, China ²College of Chemistry and Chemical Engineering, Xiamen University, Xiamen 361005, China

^{a)}Author to whom correspondence should be addressed: zhangdan@xmu.edu.cn

ABSTRACT

The relative optical gains at three near infrared wavelengths (1550 nm, 1064 nm, and 980 nm) were achieved in NaYF₄: Er^{3+} , Yb^{3+} nanoparticle-doped SU-8 waveguides when using two low-cost light-emitting diodes (LEDs) instead of traditional 980 nm semiconductor laser as pump source. The polymer waveguides were fabricated by one-step photolithography process. The fluorescence bands around 1550 nm and 1000 nm wavelengths due to the ${}^{4}\text{I}_{13/2} \rightarrow {}^{4}\text{I}_{15/2}$ transition of Er^{3+} ions and ${}^{2}\text{F}_{5/2} \rightarrow {}^{2}\text{F}_{7/2}$ transition of Yb³⁺ ions were observed under the excitation of 405 nm and 520 nm LEDs. By using the vertical top pumping mode of LEDs, the relative gains of 4.2 dB, 1.7 dB, and 2.1 dB at 1550 nm, 1064 nm, and 980 nm wavelengths were achieved, respectively, on a 10-mm-long waveguide.

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Erbium-doped waveguide amplifiers (EDWAs) have played an important role in the integrated optics on chip. They can be integrated with optical waveguide devices, such as coupler, arrayed waveguide grating, optical switch, and modulator to compensate their optical losses to form an efficient on-chip optical system.¹⁻⁵ Generally speaking, inorganic materials (silicate, phosphate, crystals, and alumina) and polymer can also be used as doping hosts of rare earth Er³⁺ ions to fabricate EDWAs. Among them, polymer EDWAs have received increasing attention due to the easily adjustable refractive index, simple fabrication processing, and low cost. Erbium ytterbium codoped fluoride nanoparticles, such as LaF₃: Er³⁺, Yb³⁺⁶ and BaYF₄: Er³⁺, Yb^{3+,7} which are doped in organic-inorganic hybrid materials or polymer materials, have become mainstream materials to fabricate polymer EDWAs over the years. A typical gain value of 4 dB/cm could be achieved.^{8–11} Usually, a pump source is needed to realize the population inversion of Er3+ions, and 980 nm semiconductor lasers are often selected because of the intrinsic absorption of Yb3+ ions (2F7/2 \rightarrow ²F_{5/2}) and Er³⁺ ions (⁴I_{15/2} \rightarrow ⁴I_{11/2}) at the 980 nm wavelength. Almost all the theoretical models of gain simulation are established under the excitation of 980 nm lasers. This idea has not been broken through for a long time. The traditional pumping method has several disadvantages. In order to obtain the optical gain, the intrinsic

absorption cross section which is on the order of 10^{-20} cm² to 10^{-21} cm² often requires a high laser pump power (100–400 mW), thus causing the energy upconversion effect of Er³⁺ ions and thermal damage of waveguides.^{12,13} Also, a wavelength division multiplexer (WDM) is needed to couple with a 980 nm laser diode (LD) and 1550 nm signal laser. It will bring additional optical losses,¹⁴ and it is difficult to reduce the commercial cost by using semiconductor lasers as pump sources.

In this work, two light-emitting diodes (LEDs) were used to pump NaYF₄: $\rm Er^{3+}$, $\rm Yb^{3+}$ nanoparticle-doped SU-8 polymer waveguides amplifier. The channel waveguides were fabricated by simple one-step photolithography process. The photoluminescence (PL) spectrum was characterized under the excitation of LEDs. By using the vertical top pumping mode of 405 nm and 520 nm LEDs, the relative gains of 4.2 dB, 1.7 dB, and 2.1 dB at 1550 nm, 1064 nm, and 980 nm wavelengths were achieved, respectively, on a 10-mm-long waveguide.

An erbium- and ytterbium-doped sodium yttrium oxyfluoride (NaY_{1.54}Yb_{0.40}Er_{0.06}F₅O) was purchased from Sigma-Aldrich Trading Co. Ltd. The molar ratio of Er^{3+} :Yb³⁺ is about 1:7. The SU-8 3005 (Kayaku Advanced Materials, Inc.) was selected as a host of nanoparticles because the photoresist properties of SU-8 can be used to fabricate waveguides by only one-step photolithography process.

The concentration of NaYF₄: Er^{3+} , Yb^{3+} nanoparticles in SU-8 is 1 wt.%.

Figure 1 shows the absorption spectrum of the NaYF₄: Er^{3+} , Yb^{3+} nanoparticle powder taken by a SHIMADZU UV-visible spectrophotometer. The absorption bands at 400–425 nm, 450 nm, 488 nm, 520 nm, 544 nm, 650 nm, 798 nm, and 970 nm wavelengths corresponded to the transitions of Er^{3+} ions from ground state ${}^{4}I_{15/2}$ to excited states ${}^{2}H_{9/2}$, ${}^{4}F_{5/2}$, ${}^{4}F_{7/2}$, ${}^{2}H_{11/2}$, ${}^{4}S_{3/2}$, ${}^{4}F_{9/2}$, ${}^{4}I_{9/2}$, and ${}^{4}I_{11/2}$. The absorption bands at 918 nm and 970 nm wavelengths also corresponded to the transitions from ground state ${}^{2}F_{7/2}$ to excited state ${}^{2}F_{5/2}$ of Yb^{3+} ions. The spectrum shows an obvious continuous absorption band in the ultraviolet region. This means that it is possible to realize the population inversion of Er^{3+} and Yb^{3+} ions under the excitation of a blue-violet LED.

A 405 nm LED was selected as pump source, and a visible light LED with a central wavelength of 520 nm, corresponding to the intrinsic absorption of Er^{3+} ions (${}^{4}\text{I}_{15/2} \rightarrow {}^{2}\text{H}_{11/2}$), was used for comparison. The photoluminescence (PL) spectrum of the NaYF₄: Er^{3+} , Yb³⁺ nanoparticle is shown in Fig. 2. The PL peaks at 1460–1580 nm and 950–1100 nm were due to the transitions from the state ${}^{4}\text{I}_{13/2} \rightarrow {}^{4}\text{I}_{15/2}$ of Er^{3+} ions and the state ${}^{2}\text{F}_{5/2} \rightarrow {}^{2}\text{F}_{7/2}$ of Yb³⁺ ions, respectively.¹⁵ The fluorescence full width at half maximum (FWHM) at 1535 nm was about 27 nm. At the same pump power, the PL intensity excited by 405 nm LED was stronger than that excited by 520 nm LED.

Using NaYF₄: Er^{3+} , Yb³⁺ nanoparticle-doped SU-8 3005 as core material, waveguides with rectangular core cross section structure were achieved. A 3 μ m thickness core layer was first spin-coated onto a silicon substrate with a 2 μ m thickness SiO₂ as bottom cladding layer. By standard photolithography, a group of waveguides with 6 μ m width and 3 μ m height were fabricated. The refractive index of the core is 1.553 at 1550 nm wavelength. Figure 3 indicates the scanning electron microscope (SEM) micrograph of the rectangular waveguide.

By using the vertical top pumping mode of the LED, an experimental setup for the optical gain measurement was established. The schematic diagram is shown in Fig. 4(a). As a contrast, the traditional measurement method of a 980 nm LD and a 1550 nm signal laser coupled by a 980/1550 nm WDM is also adopted for comparison, as shown in Fig. 4(b). The output signal was coupled into an Ocean



FIG. 1. Absorption spectrum of NaYF₄: Er^{3+} , Yb³⁺ nanoparticles.



FIG. 2. Room temperature infrared PL spectrum of NaYF₄: Er^{3+} , Yb³⁺ nanoparticles under the excitation of 405 nm and 520 nm LEDs.

Optics FLAME-NIR-INTSMA25 optical spectrometer. Through the top pumping mode of the LED, the pump power could be uniformly distributed throughout the waveguide, EDWAs can be more flexibly placed in the position of planar optical interconnection, and the addition loss caused by the use of WDM component can also be avoided.

The optical gains in the channel waveguide at 1550 nm wavelength were observed under the excitation of 405 nm and 520 nm



FIG. 3. The SEM micrograph of NaYF₄: Er^{3+} , Yb^{3+} nanoparticle-doped SU-8 rectangular waveguides.



FIG. 4. The experimental setup for optical gain of polymer waveguide amplifier: (a) the top-pumping LED mode; (b) the traditional 980 nm laser pumping mode.

LEDs. Figure 5(a) shows the output optical intensity of a 10-mm-long channel waveguide as a function of pump power under the excitation of 405 nm LED. The input signal power is 0.2 mW at 1550 nm wavelength. The signal intensity is linearly enhanced as the pump power is increasing. When the pump power reaches 77 mW and 173 mW, respectively, the output optical intensity of the waveguide increases by 30.4% and 138.9%. Figure 5(b) shows the relative gain as a function of pump power with different input signal powers. The relative gain is defined as $10lg(P_{P+S}/P_s)$, where P_{P+S} and P_s are the output signal powers as measured with and without pump power, respectively.¹⁰ It is shown that for a fixed pump power, the smaller the input signal power is, the higher the gain is. The maximum gain value increases from about 2.2 dB to 4.2 dB as the signal power varies from 0.5 to 0.2 mW for a pump power at 225 mW. The optical gain is compared when pumped by 405 nm and 520 nm LEDs, respectively, as shown in Figure 5(c). The results show that the relative gain performance of 405 nm LED is better than that of 520 nm LED, which is consistent with the PL spectrum measured in Fig. 2. One of the main reasons is that the absorption cross section of NaYF₄: Er³⁺, Yb³⁺ nanoparticlesdoped SU-8 polymer material in the ultraviolet region is much higher than that in the visible region, as shown in Fig. 1. When the 405 nm LED pump power reaches 225 mW, the maximum gain is 4.2 dB, while when the 520 nm LED pump power reaches 300 mW, the maximum gain is 2.1 dB. The optical gain pumped by 980 nm laser is also demonstrated in Fig. 5(d) by using the measurement method shown in Fig. 4(b). When the 980 nm pump power is 327 mW, the maximum gain value reaches about 2.3 dB, followed by gain saturation and decrease. This phenomenon is common in the study of polymer optical waveguide amplifier under excitation of lasers.¹⁸⁻²¹ Many factors, such as depletion of some levels of Er³⁺ ions, upconversion effects, and propagation loss, could influence the gain of the waveguide

amplifier.^{22,23} Also, the thermal damage of the waveguides is a cause of gain saturation and decrease. When a 200 mW 980 nm pump laser is input into the end of the waveguide, the incident optical power density of the waveguide's cross section can reach about 1.1×10^9 mW/cm². At this strong power density, the waveguide is easy to damage and could not work for a long time. Although the power of LED is also 200 mW, the power density of waveguide is calculated as about 100 mW/cm² when the waveguide is placed about 0.5 cm below the LED. It is much lower than that of 980 nm laser. Therefore, the trend of gain saturation is not observed in the pump power range of 0–300 mW under the excitation of LEDs, and the stability of the amplified signal is much better.

Due to the transitions of Yb³⁺ ions from the state ${}^{2}F_{5/2}$ to ${}^{2}F_{7/2}$ level, the optical gains at 980 nm and 1064 nm wavelengths were also obtained in the channel waveguides. Figure 6(a) shows the relationship between output signal intensity and pump power under 405 nm LED excitation when the input signal wavelength is 1064 nm. The relative gain vs pump power is shown in the inner illustration. The gain increases linearly with the pump power. When the pump power is 374 mW and the signal power is 0.5 mW, a maximum gain of approximately 1.7 dB at 1064 nm is observed. The gain performance at 980 nm is similar to that at 1064 nm, as shown in Fig. 6(b). A maximum gain of 2.1 dB is observed under the excitation of the 173 mW 405 nm LED. Under the excitation of 405 nm LED, there are a few reports about the corresponding intrinsic absorption state of Yb³⁺ ions.²⁴⁻²⁶ We consider that there is a process of energy transfer between host material (NaYF4 nanoparticles and polymer SU-8) with Yb³⁺ and Er³⁺ ions. Similar energy transfer mechanism also occurs between Er³⁺ ions and Si nanocrystals,²⁷ as well as between organic ligands and central Nd³⁺ ions.²⁸ When the host material, especially polymer SU-8, absorbed blue violet light over a broad wavelength range (200–450 nm) and coupled with Yb^{3+} ions, these ions can be excited from the ground state ${}^{2}F_{7/2}$ to ${}^{2}F_{5/2}$ state by energy transfer and then achieve 980 nm and 1064 nm emissions. Meanwhile, backward energy transfer process between ${\rm Er}^{3+}$ and Yb^{3+} ions, ${}^{4}I_{11/2}$ + ${}^{2}F_{7/2}$ \rightarrow $^2F_{5/2}+$ $^4I_{15/2},$ can also cause the emission of Yb $^{3+}$ ions. In addition to the intrinsic absorption of Er^{3+} ions from ${}^{4}I_{15/2}$ to ${}^{2}H_{9/2}$ state corresponding to 405 nm, there is also an energy transfer process between the host material and Er³⁺ ions. Both of them can help Er³⁺ ions achieve 1550 nm stimulated emission, as shown in Fig. 7. Therefore, the optical amplification of 1550 nm, 980 nm, and 1064 nm wavelengths by Er³⁺ and Yb³⁺ ions transitions can be obtained simultaneously under 405 LED pumping.

In conclusion, the relative optical gains at near infrared wavelengths were demonstrated in NaYF₄: Er^{3+} , Yb^{3+} nanoparticle-doped SU-8 waveguides. By using the vertical top pumping mode of 405 nm and 520 nm LEDs, optical gains of 4.2 dB and 2.1 dB were obtained in a 10 mm-long waveguide when the input signal power was 0.2 mW at 1550 nm. The relative gains of 2.1 dB at 980 nm and 1.7 dB at 1064 nm wavelengths were also obtained in the channel waveguides due to the transitions of Yb³⁺ ions from the state ${}^{2}F_{5/2}$ to ${}^{2}F_{7/2}$ level. A process of energy transfer between Er^{3+} , Yb³⁺ ions and polymer host under 405 nm LED pumping was discussed. The results show that the polymer EDWA pumped by LED could achieve similar optical gain to that pumped by 980 nm laser. The LED top-pumping mode is more stable, convenient, and low-cost, and could avoid the disadvantages of thermal damage of waveguides and addition losses caused by high-power



FIG. 5. (a) The output signal intensity as a function of pump power (under 405 nm LED excitation). (b) The relative gain as a function of pump power with different input signal power. (c) Comparison of optical gain with 405 and 520 LEDs pumping (signal power is 0.2 mW). (d) The output signal intensity and relative gain (inset) as a function of pump power (under 980 nm laser excitation).

laser pumping and the use of WDM components. In addition, it is possible to achieve electroluminescence by doping NaYF4: Er^{3+} , Yb³⁺ nanoparticles into some host materials of bipolar transmission. Holes and electrons can be injected into the bipolar host from the anode and cathode, respectively, and then transmitted directly in the host. When they combine to form excitons and transfer energy to nanoparticles, luminescence can be achieved.^{29,30} The operation bandwidth of the EDWA could be further widened by optimizing the ratio of Er^{3+} and Yb³⁺ ions



FIG. 6. The optical amplification at (a) 1064 nm and (b) 980 nm by 405 nm LED pumping.

and increasing the cross section of waveguide.³¹ Also, the polymer EDWA with LED as pump source has the potential to be waveguide lasers by depositing distributed Bragg reflector (DBR) on the end faces of the waveguide and fabricating Bragg gratings on the chip.^{32,33} It is expected to play an important role in optical sensing³⁴ and optical interconnects.³⁵



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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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