Optical gain of LaF_3:Nd nanoparticle doped polymers for active integrated optical devices.

R. Dekker[1)], J.W. Stouwdam[2)], D.J.W. Klunder[3)], A. Borreman[4)], M.B.J. Diemeer[1)], K. Worhoff[1)] and A. Driessen[1)].

[1)]University of Twente, Faculty of Electrical Engineering, Mathematics and Computer Science, P.O.Box 217, 7500 AE Enschede, The Netherlands. Phone: +31-53-489 4440; E-mail: R.Dekker@utwente.nl.

[2)]University of Twente, Faculty of Science and Technology, P.O.Box 217, 7500 AE Enschede, The Netherlands.

[3)]C2V, P.O.Box 318, 7500AE Enschede, The Netherlands.

[4)]LioniX BV, P.O.Box 456, 7500AH Enschede, The Netherlands.

We report on rare earth doped LaF_3 nanoparticles dispersed in PMMA and SU-8 photosensitive polymers. We observed optical gain after we applied these materials for waveguides. Experimental results on various samples will be discussed. We theoretically discuss the improvements that can be obtained and the possibilities of this new class of materials when applied to microring resonators.

Introduction

Over the past few years, more and more new high-tech materials are emerging with very promising properties for use in active integrated optical devices. By combining the broad range of polymer properties and the long lifetime of rare earth dopants in inorganic nanoparticles, an enormous amount of flexibility regarding material properties can be achieved. In this article, the rate equations for the absorption and emission characteristics of LaF_3:Nd around 1330nm will be explained first, followed by a description of our pump experiments on LaF_3:Nd doped polymer waveguides. Stimulated emission and gain are demonstrated, which is promising for application in active integrated optical microring resonator devices. We conclude with some improvements and plans for the near future.

Theory

Theoretically Nd^{3+} can be described as a four level system as shown in Figure 1. The Nd^{3+} ions are excited from the $^4I_{9/2}$ ground state to the $^4G_{5/2}$ excited state followed by a fast decay to the $^4F_{3/2}$ excited state. At the 1330nm emission in the second telecom window, the lower level is the $^4I_{13/2}$ level, which decays rapidly to the $^4I_{9/2}$ ground state. Due to the fast decays 5→4 and 3→1, the levels 5 and 3 are considered empty and the populations of the levels 4 and 1 can be determined as follows[1]:

$$\frac{dN_1}{dt} = 0 = -W_{15}N_1 + \left(\frac{1}{\tau} + W_{43}\right)N_4 \qquad \frac{dN_4}{dt} = 0 = +W_{15}N_1 - \left(\frac{1}{\tau} + W_{43}\right)N_4 \qquad N_1 + N_4 = N_{tot}$$

which are equal to zero in the steady state. Because the sum of both populations equals the total population, this set of equations simplifies to:

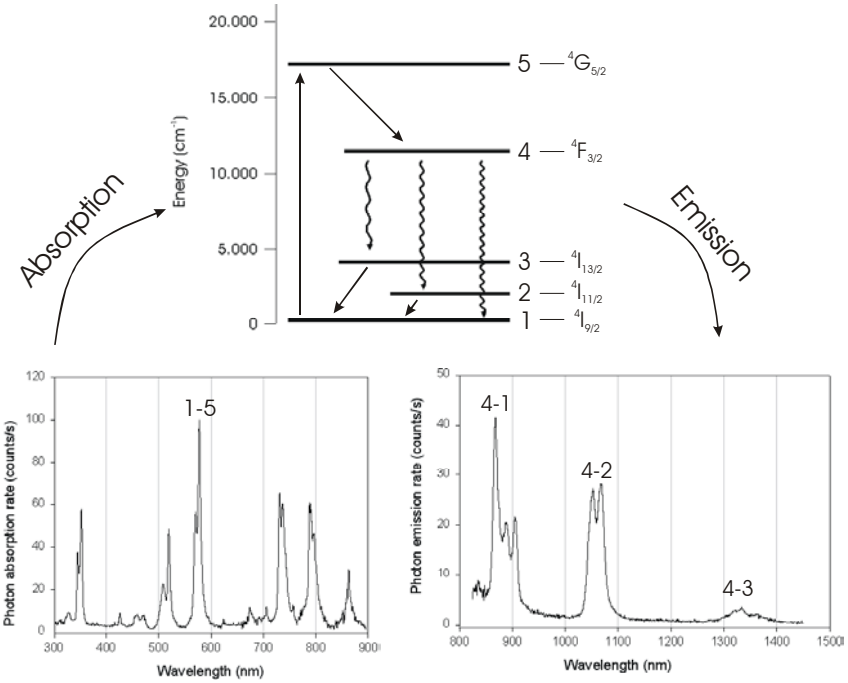

Figure 1: Energy diagram, absorption spectrum and emission spectrum of LaF_3:Nd in solution. 1-5 *denotes the absorption near 575nm, whereas 4-1, 4-2 and 4-3 represent the emission around 880, 1050 and 1330 nm respectively.*

$$N_1 = \left(\frac{W_{43} + 1/\tau}{W_{43} + 1/\tau + W_{15}}\right)N \qquad N_4 = \left(\frac{W_{15}}{W_{43} + 1/\tau + W_{15}}\right)N$$

The absorption and emission rates W_{15} and W_{43} are defined as follows:

$$W_{15} = \sigma_a I_p \; ; I_p = \frac{P_p}{h\upsilon_p A} \qquad W_{43} = \sigma_e I_s \; ; I_s = \frac{P_s}{h\upsilon_s A}$$

where I_p and I_s are the photon fluxes of pump and signal source respectively and σ_a the absorption cross section and σ_e the stimulated emission cross section. The photon fluxes are calculated from the pump and signal power P_p and P_s, the light frequencies ν_p and ν_s, Planck's constant h and the cross section of the waveguide A. The power of the signal and pump along the waveguide is given by:

$$dI_s = W_{43} N_4 dz \qquad dI_p = W_{15} N_1 dz$$

We will use these equations to predict and validate our gain measurements.

Experimental

For our experiments we used LaF_3:Nd nanoparticles that are composed of LaF_3-crystals where ~5 % of the lanthanum ions are replaced by neodymium ions. By choosing LaF_3 as host for the Nd^{3+} ions, quenching will be reduced to a minimum because of the very low vibrational energies of LaF_3 [2]. In order to prevent clustering of the nanoparticles, which will cause light scattering, and to control the growth of the particles during the synthesis, ligands (which add up to about 10% of the mass) are attached to the nanoparticles. Figure 2 shows a schematic representation and transmission electron microscope (TEM) images of the LaF_3:Nd nanoparticles.

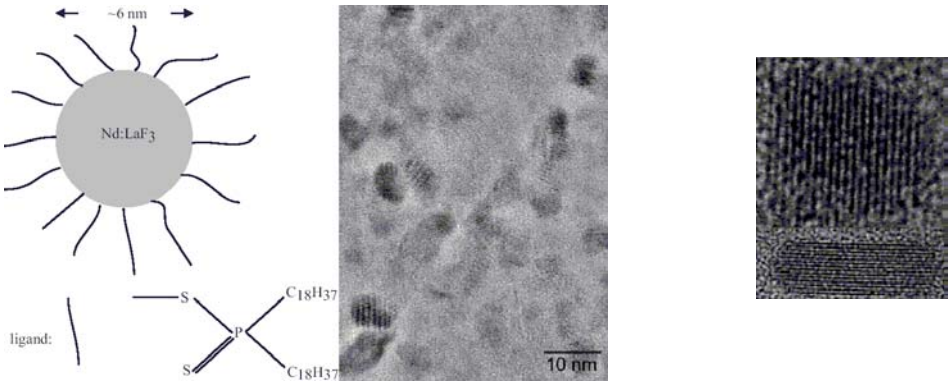

Figure 2: Left - schematic representation of the nanoparticles and its ligands; Middle – TEM image of the nanoparticles; Right: TEM zoom clearly showing the crystalline nature of the nanoparticles.

For our first sample we spinned a PMMA solution with 10 weight percent LaF_3:Nd onto a thick SiO_2 buffer (for details on preparation see Kuiper[3]). After patterning with standard photoresist and reactive ion etching of the 3.3µm thick doped PMMA film, we obtained straight waveguides with a channelwidth of 10µm and a ridgeheight of 1.4µm[1]. A second sample was made using SU-8 photoresist as the host for the nanoparticles (20 weight percent). Waveguides with a width of 10µm were fabricated by UV-exposure of the 10µm thick doped SU-8 film followed by a standard photoresist development step[1]. A third sample consisted of a Si_3N_4 straight monomodal high contrast waveguide with a 20 weight percent nanoparticle doped PMMA cladding on top [3,4].

Measurements

A signal (λ_s=1319nm) and a 12mW chopped pump (λ_p=578nm) beam were coupled into the PMMA waveguides. Spontaneous emission signals were ~30dB lower than the signal power, not interfering with our amplification measurements. The signal wavelength was fed into a detector after filtering out the other wavelengths. A signal enhancement of 0.3 dB in a 3cm long waveguide has been observed. Numerically solving the rate equations outlined above using the values for the different parameters shown in Table 1 will yield the solid line presented in Figure 3, which agrees well with our measurements.

Table 1:Parameters used in the optical gain calculations.

Parameter	Value	Unit
P_p	12	mW
P_s	1	mW
σ_a	2.5×10^{-20}	cm^2
σ_e	5×10^{-21}	cm^2
λ_s	1319	nm
λ_p	578	nm
τ	200	µs
N_{tot}	1.4×10^{19}	cm^{-3}
Height	3.3	µm
Width	10	µm

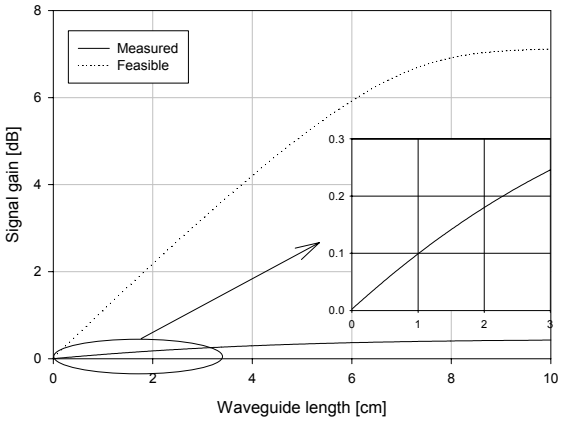

Figure 3: Gain vs length plots. Solid line represents our experiments. Dotted line represents theoretically feasible gain/cm.

A similar experiment yielded a small 0.1dB signal gain on our second sample, a 5.2cm SU-8 waveguide with only 5mW of pump power. The pump wavelength had to be changed from 578nm to 795nm due to the high background absorption of SU-8.

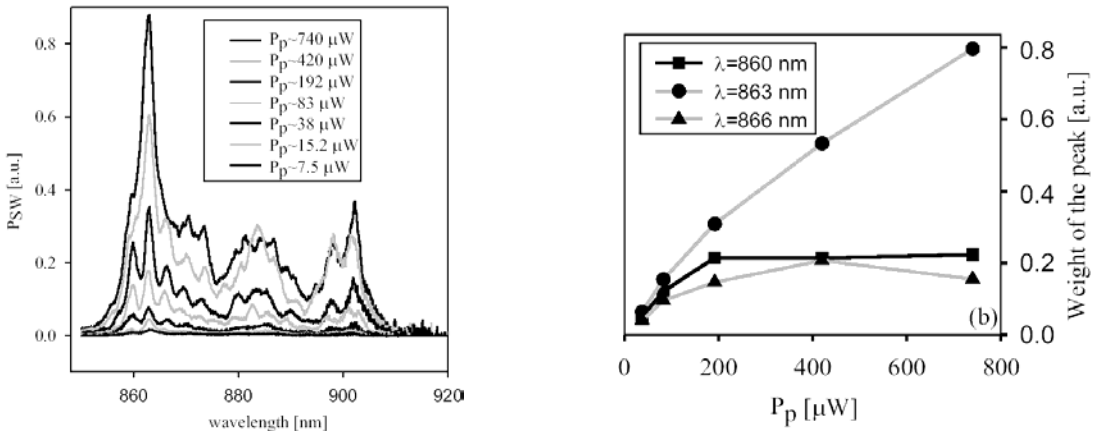

Figure 4: Left – emission versus pump power for a LaF_3:Nd doped PMMA straight waveguide; Right – relative weight of emission peaks versus pump power.

The emission spectrum of our Si_3N_4 waveguide with nanoparticle doped PMMA cladding was measured at different pump powers as shown in Figure 4. The emission at 863nm monotonously increases with increasing pump power, while the emission peaks at 860nm and 866nm show saturation at P>192μW. This might be an indication that stimulated emission at 863nm is taking place in this sample[4].

Future applications and improvements

Although the observed amplification in our high loss multimode waveguides is still very low, we believe that we can easily improve the performance by changing our technology and design. For instance, the dotted line in Figure 3 shows the theoretically feasible gain for a 4×4μm PMMA waveguide with 40 weight percent of nanoparticles and 100mW of pump power. We are currently working on high index contrast, highly confined LaF_3:Nd nanoparticle doped microring resonators in which the power buildup in the ring will further enhance the input pump power. As the nanoparticle host we are investigating state of the art photosensitive bromated polymer materials with very low background losses in the visible and infrared. Erbium doped nanoparticles have been synthesized as well for use in the third telecom window. Our aim is to build microring lasers and all-optical switches utilizing the gain and the third-order nonlinear properties of this new class of compound materials.

References

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