Neodymium doped lead lanthanum zirconate titanate ferroelectric transparent ceramics as a potential mode-locked laser

Andrea Simone Stucchi de Camargo,¹,² Luiz Antonio de Oliveira Nunes,¹
Ériton Rodrigo Botero,² Ducinei Garcia,² José Antonio Eiras²
¹Instituto de Física de São Carlos – Universidade de São Paulo
²Departamento de Física – Universidade Federal de São Paulo
andreasc@if.sc.usp.br

Abstract

Neodymium doped transparent lead lanthanum zirconate titanate bulk ceramic has shown to be a promising laser active medium at 1.06 µm. Given the ferroelectric nature of the PLZT host, efficient intensity modulation of Nd³⁺ emissions can be achieved by application of an external electrical field to the samples. This work presents luminescence spectra and excited state lifetime data, which suggest that Nd:PLZT could be efficiently used as an integrated mode-locked laser.

Introduction

Neodymium doped transparent ceramics have recently attracted much attention as near-infrared laser active media at 1.06 µm (⁴F₃/₂ → ⁴I₁₁/₂), due to the many advantageous characteristics they offer in comparison to crystals and glasses. Since ceramics offer higher flexibility in tailoring optical properties and are generally able to incorporate higher concentrations of doping ions, they are especially interesting for high power lasers [1-3]. In that sense, the composition that has been mostly investigated is Nd:YAG [2,3]. However, it has been shown that ion-ion energy transfer processes introduce a strong self-quenching of fluorescence quantum efficiency in this ceramic for concentrations higher than 3.5 × 10²⁰ ions/cm³ [2]. Therefore, the search for ceramic hosts that are less sensitive to concentration quenching is justified.

In recent years we have been investigating highly transparent Nd³⁺ doped lead lanthanum zirconate titanate (PLZT) transparent ceramics, which present desirable characteristics of an efficient laser active medium. Among these characteristics are an extensive transmission window (up to 7.5 µm), low phonon energy (750 cm⁻¹), absence of OH⁻ groups, fairly high absorption and emission cross sections (σ⁸⁰³ nm = 1.5 × 10⁻²⁰ cm² and σ¹⁰⁶ µm = 3.5 × 10⁻²⁰ cm²), negligible excited state absorption losses around 1.06 µm [4], and very low probability of downconversion and upconversion energy transfer losses, leading to a quenching factor Q = 14.5 × 10²⁰ ions/cm³ [5]. But the most interesting feature of this material is the possibility of introducing a modulation effect of its 1.06 µm emission by application of an external electrical field in the own active medium.

In this work we present a detailed discussion on the spectroscopic characteristics of this new transparent ceramic material, as well as on the results obtained from modulated luminescent and excited state lifetime experiments.
Experimental Setup

The Nd:PLZT transparent ceramic, doped with $1.0 \text{ wt\% Nd}_2\text{O}_3$ ($2.8 \times 10^{20}$ ions/cm$^3$), studied in this work, was obtained with ~1.5 mm thickness, through conventional solid state reaction followed by uniaxial hot pressing at 1250 °C for 3 h [6]. The modulated luminescence experiments were carried out by excitation of the sample with a diode laser at 800 nm and by varying the electrical field, applied to the sample, up to 750 V. The signal was collected by an InGaAs detector. Excited state lifetime values were obtained from the luminescence decay curves in time which were measured with excitation from a Nd:YAG pumped OPO laser, by eliminating the monochromator from the mounting, and using a color filter and a large detector, instead.

Results and Discussions

Figure 1 presents the luminescence spectra of Nd:PLZT (1.0 wt% Nd$_2$O$_3$) as a function of the applied voltage. As it can be seen, a significant increase in intensity of the bands from 0.85 to 1.5 µm is observed with increasing electrical field intensity. The inset presents the integrated area of 1.06 µm band, also as a function of the applied voltage. The linear dependence suggested that the modulation is due to the piezoelectric effect that causes a misalignment of the sample as it vibrates with different frequencies. This assumption was confirmed by experiments from which the variation in position of a transmitted HeNe laser beamspot is clearly seen in the far field. It is worth noting that in the spectra, no change in spectral lineshape or width due to additional Stark splitting or energy dislocation of electronic levels is observed, by application of the electrical field. This is an indication that no significant alteration of ligand field around the doping rare earth ion is taking place.

![Figure 1: Luminescence intensity modulation as a function of electrical field strength for a Nd:PLZT (1.0 wt% Nd$_2$O$_3$) sample with 1.5 mm thickness.](image)

This observation is corroborated by analyzing the lifetime values of level $^4F_{3/2}$ as a function of doping concentration. Figure 2 presents the decay curves in logarithmic scale from which the lifetimes were obtained. The values are exactly the same in the presence or absence of the electrical field and since the lifetime is inversely proportional to transition probabilities, and thus very sensitive to alterations in chemical bonds, it is confirmed that the applied electrical field does not affect the local environment around Nd$^{3+}$ ions significantly. The reason for this lies in the fact that intraconfigurational 4f-4f transitions in trivalent rare earth ions are screened by more external 5s and 5d orbitals, which make these ions less sensitive to ligand field changes.

![Figure 2: Decay curves in logarithmic scale from which the lifetimes were obtained.](image)
These results associated with the desirable characteristics of an efficient laser material exhibited by transparent Nd:PLZT ceramics, suggest that this innovative material, can be used in the bulk or thin film form for the construction of not only a mode-locked laser, in which both laser action and modulation would be done in the same active medium, but also of other multifunctional devices like waveguides and sensors, for instance.

![Image of excited state lifetime values as a function of concentration and under a 750 V external electrical field for Nd:PLZT (0.1 – 4.0 wt% Nd$_2$O$_3$) ceramics.]

**Figure 2:** Excited state lifetime values as a function of concentration and under a 750 V external electrical field for Nd:PLZT (0.1 – 4.0 wt% Nd$_2$O$_3$) ceramics.

**Conclusions**

It is concluded that efficient intensity modulation of 1.06 µm emission in Nd:PLZT transparent ceramics can be achieved by application of an external electrical field in the sample. This result shows good prospects for the construction of compact, low cost multifunctional devices, such as a mode-locked laser, where the gain losses are introduced in the own laser active medium.

**Acknowledgements**

The authors thank FAPESP and CNPq for the financial support of this work.

**References**


