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Excited state spectroscopy and laser properties of the Nd³⁺ doped LaBGeO₅ single crystal

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ABSTRACT

The excited state excitation spectra of Nd³⁺ in LaBGeO₅ crystal have been systematically investigated in the 1.048 μm and 0.54 μm wavelength spectral domains. First cw laser experiments under Ti-Sapphire laser pumping are reported.

INTRODUCTION

LaBGeO₅:Nd³⁺ is a promising nonlinear laser crystal for applications in integrated optics. It offers some advantages over the more developed LiNbO₃:MgO:Nd³⁺ minilasers [1], such as absence of photorefractive damage, no domain structure and high distribution coefficient for the lasing Nd³⁺ ions. Its laser line at 1.048 μm is particularly suitable for obtention of 0.524 μm laser radiation via self-frequency doubling (SFD) [2]. Consequently a detailed study of the optical properties around these two spectral regions becomes necessary.

In previous papers [2,3] the absorption, luminescence and second harmonic generation properties of this material have been reported. In this work the excited state absorption (ESA) spectra have been systematically investigated in the wavelength domains around 1.048 μm and 0.524 μm (SFD), via excited-state excitation (ESE) [4]. In addition, first cw Ti-sapphire pumped laser experiments have been performed to simulate operation under diode-laser pumping.

EXPERIMENTAL

Single crystals of LaBGeO₅ doped with 1.4 % of Nd³⁺ ions were used. Experimental details about ESE experiments can be found in ref.[4]. Laser experiments were performed by pumping with a 900 mW cw Ti sapphire laser.

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RESULTS

Figure 1 is a simplified energy diagram of Nd³⁺ doped LaBGeO₅ crystal, showing the energy gap among the different states. The width of each state indicates its crystalline field splitting. This diagram allows for identification of the fluorescence lines reported in this work with transitions between defined states. Upwards transitions involving the main ESA processes investigated in this work are indicated by arrows.

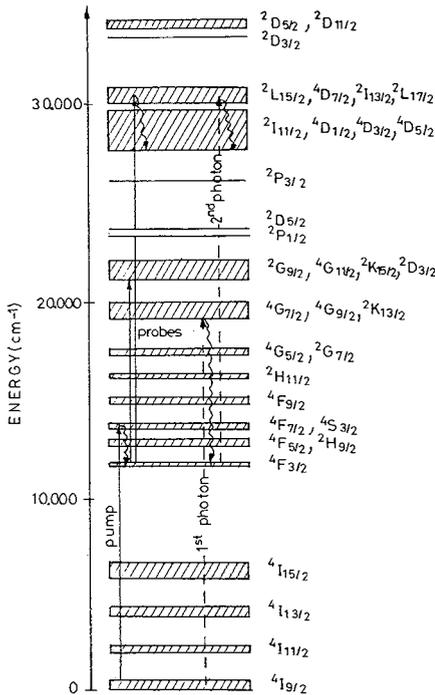


Figure 1

These ESE spectra can now be calibrated in units of ESA cross sections as reported in [3]. First of all, we estimate the Judd-Ofelt parameters from our RT polarized absorption spectra ; $\Omega_2 = 1.8 \times 10^{-20} \text{ cm}^{-2}$, $\Omega_4 = 3.5 \times 10^{-20} \text{ cm}^2$ and $\Omega_6 = 4 \times 10^{-20} \text{ cm}^2$. After that we calculate the integrated $4F_{3/2} \rightarrow 2G_{9/2}$ ESA cross section, $\int \sigma d\lambda = 8.57 \times 10^{-20} \text{ cm}^3$, and then we consider that it is equal to our integrated ESE spectrum, $\int I d\lambda$, so obtaining the peak cross section values; $\sigma_{\perp} = 2 \times 10^{-20} \text{ cm}^2$ and $\sigma_{\parallel} = 9 \times 10^{-21} \text{ cm}^2$. In every case the

The room temperature (RT) unpolarized emission spectrum under uv excitation (355 nm) shows structured emission bands around 360, 386, 420 and 458 nm. These emissions correspond to different inter-Stark transitions from the high lying $4D_{3/2}$ and $2P_{3/2}$ states to the $4I_J$ manifold. Excitation with visible light also produces weaker emissions departing from the $4G_{7/2}$ levels. The ESE spectra of all these emissions can be used to obtain the ESA at different wavelength domains [4]. These ESE spectra were taken after pumping with two beams: a "pump beam", to populate the excited state $4F_{3/2}$ from which ESA is originated, and the "probe beam" to scan the excited state excitation (full arrows on Fig. 1). Figure 2a shows the RT polarized ESE spectra in the wavelength domain around the 1.048 μm laser line.

This spectral region is associated with the $4F_{3/2} \rightarrow 2G_{9/2}$ intermanifold transition

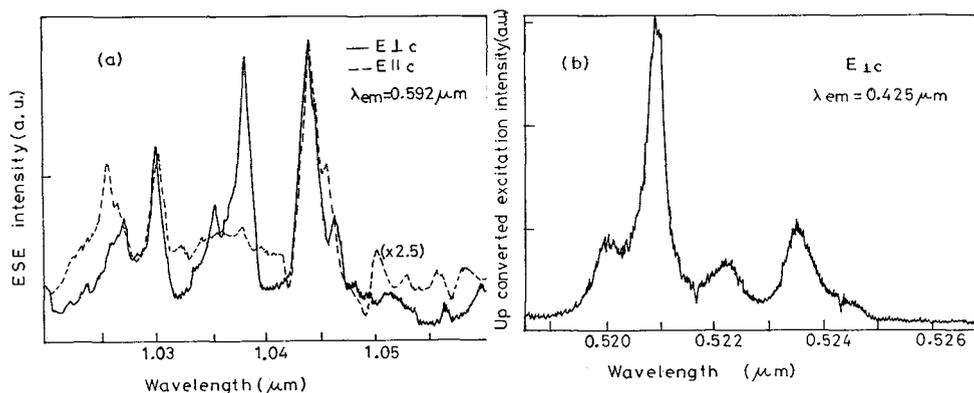


Figure 2

estimated ESA at the laser wavelength $1.048 \mu\text{m}$ is very low ($< 2 \times 10^{-23} \text{ cm}^2$) if compared with the emission peak cross section at this wavelength, $2.4 \times 10^{-19} \text{ cm}^2$ [3], so that no significant losses are expected to occur by this channel.

The ESA in the spectral region around $0.524 \mu\text{m}$ (SFD) has also been investigated. ESA mainly occurs as a second step of a green to blue up-conversion process (dotted arrows on Fig.1). This process involves a two-step two-photon absorption from the ground state $^4I_{9/2}$. The first photon populates the $^4F_{3/2}$ metastable state after non-radiative relaxation from the $^4G_{7/2}$ state. Then a second photon populates the $^4D_{3/2}$ state (after non-radiative relaxation from the upper states, see Fig.1) and the set of visible emissions mentioned above is observed. Fig. 2b shows the excitation spectrum of this up-converted process. This spectrum is essentially similar to the ground state absorption (GSA) spectrum and practically the same as that obtained in a two beam (infrared to populate and green to probe, full arrows on Fig.1) experiment. This indicates that the direct excited state absorption from the $^4F_{3/2}$ state in the green domain is not relevant compared with the absorption process leading to the green to blue up-conversion process. In any case the absorption (ESE and GSA) at the $0.524 \mu\text{m}$ wavelength (SFD) does not seem significant.

Finally first cw Ti-sapphire pumped laser experiments have been performed to simulate the operation of $\text{LaBGeO}_5\text{:Nd}$ under diode laser pumping. We have used an uncoated 28 mm long rod, end pumped along its c axis in a nearly

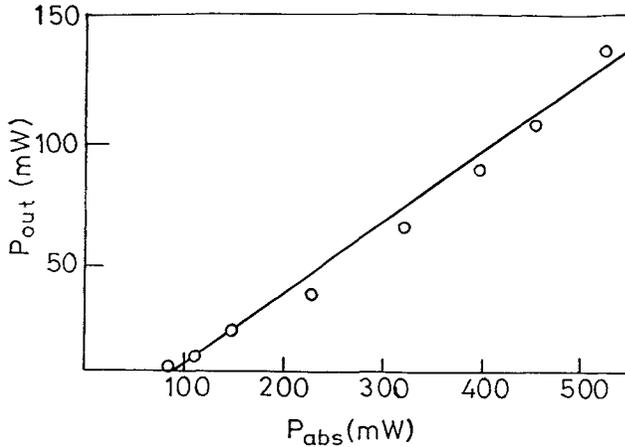


Figure 3

hemispherical cavity consisting of a flat dichroic input mirror (highly reflective around $1.05 \mu\text{m}$ ($R \geq 99.7\%$) and highly transmissive around 800 nm) and an output coupler (10 cm radius of curvature and 2% transmission around $1.05 \mu\text{m}$). Under these non-optimal conditions we have obtained (see Fig.3) a 32.6% slope efficiency and a threshold absorbed pump power of 80 mW for the $1.048 \mu\text{m}$ laser line (ELC). Further laser experiments are now being performed to improve these values.

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