



Effect of temperature on optical properties and thermal conductivity of vanadate crystals doped with thulium and erbium



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ABSTRACT

Influence of temperature on spectroscopic and thermal properties of Tm^{3+} and Er^{3+} -doped GdVO_4 and LuVO_4 crystals was examined. Polarized absorption and emission spectra as well as excited state luminescence lifetimes have been measured and analyzed especially at cryogenic temperatures in the 1.5–1.6 μm and 1.8–1.9 μm spectral ranges to assess laser qualities of these materials. The emission cross sections for $^4\text{I}_{13/2} \rightarrow ^4\text{I}_{15/2}$ (Er) and $^3\text{F}_4 \rightarrow ^3\text{H}_6$ (Tm) transitions at $T = 77\text{ K}$ were determined and confronted to the results obtained at room temperature. Effective emission cross sections estimated for aforementioned erbium and thulium transitions indicate that effective laser operation at cryogenic temperatures can be achieved in the gadolinium and lutetium vanadate crystals. Efficiency of laser material depends on its ability to heat distribution upon high-power energy pumping process. Accordingly, it has been found that the anisotropy of thermal conductivity for the undoped crystals is weakly influenced by the temperature albeit conductivity values for the GdVO_4 are higher than those for LuVO_4 at all temperatures. Incorporation of erbium or thulium ions results in a decrease of thermal conductivity in the two vanadate crystals and an anomalous temperature dependence of thermal conductivity in the LuVO_4 was observed. Anisotropy of temperature dependence of thermal conductivity was found for the $\text{LuVO}_4:3.2\text{ at}\% \text{Tm}^{3+}$ crystal. In this case the thermal conductivity along c-axis is inferior to that along a-axis of the crystal.

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1. Introduction

For many years the solid state lasers become inviting subject of study due to intensification of works on more compact, durably or sophisticated optical systems. Especially, the mid-infrared solid state lasers attract great attention because of wide area of their applications and dynamic rise of telecommunication industry requirements. The efficient oxide and fluoride laser materials have been doped with numerous optical activators but Er^{3+} and Tm^{3+} rare earths ions have been considered in particular. Erbium and thulium optical centers in crystalline hosts are characterized by the adequate schemes of energy levels. As result of that effective pump process and consequently near infrared laser generation may take place. It is known that to obtain high laser performance of optical material the emission ions should be accommodated in matrix which meets many rigorous conditions associated with specific mechanical, thermal and spectroscopic properties. Our former study implied that the erbium and thulium doped vanadate GdVO_4

and LuVO_4 crystals are appropriate materials to achieve efficient near-infrared high-power laser operation [1–5]. Laser potential of $\text{GdVO}_4:\text{Er}$ crystal obtained by flux technique was evaluated as result of spectroscopic investigation by Bertini et al. [6]. Subsequently, our optical study of Czochralski grown the erbium-doped gadolinium vanadate crystal performed in wide range of temperatures were reported [4]. Employing previously reported valuable spectroscopic parameters the diode pumped $\text{GdVO}_4:\text{Er}^{3+}$ lasers were examined [7]. Next, N. Ter-Gabrielyan et al. confirmed that $\text{GdVO}_4:\text{Er}$ crystal is efficient laser material operating at room and cryogenic temperature [8,9]. Investigation on $\text{LuVO}_4:\text{Er}^{3+}$ fundamental optical properties has been reported by Lisiecki et al. in Applied Physics B: Optics and Lasers [5]. This study has been recently complemented [10] and furthermore, first report on cryogenic resonantly-pumped $\text{LuVO}_4:\text{Er}^{3+}$ laser was documented. Spectroscopic properties of the thulium-doped vanadate crystals family were investigated in details [1]. Moreover, cw laser operation in the $\text{GdVO}_4:\text{Tm}^{3+}$ with a slope efficiency exceeding 39% and a quasi-cw mode laser oscillation in the $\text{LuVO}_4:\text{Tm}^{3+}$ were there demonstrated. Afterwards, comparison of spectroscopic features relevant to the laser performance of thulium-doped YVO_4 , GdVO_4 ,

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and LuVO₄ crystals at temperatures likely to be encountered during high power laser operation was reported [2]. Actually, since 1995 the study on the GdVO₄:Tm lasing characteristics were reported in several articles [11–13]. The first investigations of LuVO₄:Tm laser operation at room temperature were published in 2006 [14,15] and up to now limited number of papers related to this laser material is available [16,17].

A high efficiency cryogenic diode-pumped GdVO₄:5 at.% Tm,0.5 at.%Ho laser was described by Yao Bao-Quan et al. The maximum output power 4.2 W with slope efficiency of 38% at 2.05 μm was reached [18]. These authors developed the cryogenic vanadate lasers and several years later they reported a 22.3 W CW diode-pumped cryogenic GdVO₄:5 at.%Tm,0.5 at.%Ho laser [19]. At present in literature is no report on cryogenic LuVO₄:Tm laser.

Numerous physicochemical properties of vanadate crystals are responsible for their laser performances. Among others, thermal conductivity plays a crucial role in a heat flow through a laser medium during high-power pump process. Several reports have been devoted to the assessment of undoped and rare earth doped GdVO₄ and LuVO₄ thermal properties. Unfortunately, description of these vanadate crystal qualities is inconsistent, disordered and still far from satisfactory. Thermal conductivity of GdVO₄:Nd and GdVO₄:Tm crystals have been reported in Refs. [20–22]. Literature on thermal properties of rare earth-doped LuVO₄ crystals comprises reports dealing with LuVO₄:Nd³⁺ and LuVO₄:Yb³⁺ systems [23,24] but the thermal properties of GdVO₄:Er³⁺, LuVO₄:LuVO₄:Er³⁺ and LuVO₄:Tm³⁺ were not investigated, as far as we know. In works mentioned above different experimental techniques have been employed to determine thermal conductivity/diffusivity, including a flash method [21,24], a steady-state longitudinal heat-flow method [20,22] and the method of temperature wave analysis (TWA) [23]. A high discrepancy in thermal conductivity values published thus far pointing at the experimental uncertainty inherent to a particular method. For instance, large discrepancy of the reported values of GdVO₄:Nd thermal conductivity occurs that are pointed from 6.5 W/mK to 11.8 W/mK [25]. In this work we deal with GdVO₄ and LuVO₄ crystals doped with thulium and erbium that emerge as promising cryogenic laser active media. Detailed knowledge on spectroscopic and thermal features of these laser active media is crucial for the tailoring and optimization of their laser performance.

2. Experimental details

Single crystals of the GdVO₄ and LuVO₄ undoped and containing of 0.7 at.% of Er³⁺ and 3.2 at.% of Tm³⁺ were grown by the Czochralski technique. The crystals were appropriately cut and polished. In the form of cuboids the crystals were studied utilizing the optical spectroscopy methods. The concentration of optical admixtures was verified using an inductively coupled plasma (ICP-ES) measurement. Polarized absorption spectra at 77 K and 300 K have been recorded with a Varian 5E UV-VIS-IR spectrophotometer. Polarized emission spectra in the near infrared have been recorded using a setup consisting of Dongwoo Optron DM711 emission monochromator with 750 mm focal length and the InGaAs or PbS detectors depending on the spectral region. Emission spectra were excited by an Apollo Instruments diode laser emitting continuous wave (CW) radiation at 980 nm with a maximum power up to 3 W and an AlGaAs diode laser emitting 800 nm with highest available power of 1 W. Luminescence decay curves were recorded upon selective excitation provided by a Continuum Surelite optical parametric oscillator (OPO) pumped with the third harmonic of Nd:YAG. The resulting decays were attained with a cooled InSb Janson J10D detector connected to a Tektronix Model TDS 3052 digital oscilloscope. For low-temperature measurements, the

crystals were mounted in an Oxford model CF 1204 continuous-flow helium cryostat equipped with a temperature controller. Thermal conductivity and specific heat for the undoped and Er³⁺, Tm³⁺ doped vanadate crystals were investigated in the temperature region T = 50 K–300 K. Thermal conductivity was measured by the steady-state longitudinal heat-flow method. For this measurement the crystal samples in the form of rods 10 mm long and several mm² in the cross section with rod axes parallel to the c-axis (optical axis) or to the a-axis of the crystal were mounted in a liquid helium cryostat equipped with a temperature controller. The temperature drop along samples did not exceed 0.2 K and the extreme care was taken to eliminate parasitic heat flow between the samples and their surroundings. Measurements of the specific heat were performed in the 50 K–300 K temperature region with a Physical Properties Measuring System (PPMS Quantum Design).

3. Results and discussion

3.1. Spectroscopic study

High efficient ⁴I_{13/2} → ⁴I_{15/2} laser action in the resonantly pumped LuVO₄:Er³⁺ crystal has been recently found. Laser operating at room temperature at 1609 nm (CW ~ 4.6 W) with slope efficiency of 64% is reported in Ref. [10]. Not long before that, the effective room temperature resonantly pumped the GdVO₄:Er³⁺ laser was demonstrated [8]. For this material the maximum CW laser output power of 3.5 W at 1598.5 nm with slope efficiency of 56% was obtained with the Er-fiber laser pumping at 1538.6 nm. It is worth noticing that the slope efficiency of the GdVO₄:Er³⁺ laser increased to 84% at 1598.7 with pumping at 1538.6 nm when the laser cavity was properly cryo-cooled. Lowering the temperature to T = 77 K leads to increase efficiency of CW erbium laser emission and output power reached high value of 10.3 W [9]. The listed results indicate that 1.6 μm erbium laser emission in vanadate crystals can be successfully gained at cryogenic temperatures utilizing resonant pumping process into the adequate crystal field components of the higher laser level. This approach requires a strictly specific excitation wavelength and furthermore the limitations related to excitation power occur. Complex structure of erbium energy levels made it possible to pump of laser medium applying the alternative excitation sources operating in NIR spectral ranges. Our past spectroscopic study on erbium doped vanadate crystals showed that especially ⁴I_{15/2} → ⁴I_{11/2} Er³⁺ absorption around 980 nm is favorable to sufficient pump with the efficient InGaAs laser diodes. This excitation process realized at cryogenic temperature may cause effective erbium laser emission when spectroscopic and thermal features of the vanadate crystals will be beneficial. Fig. 1 presents polarized absorption and emission spectra of the GdVO₄:0.7 at.% Er³⁺ crystal measured at T = 77 K which are attributed to ⁴I_{15/2} ↔ ⁴I_{13/2} transitions of erbium. Absorption cross section was calculated according to the relation: σ_{abs}(ν) = α(ν)/N where α(ν) is absorption coefficient of this optical system and N represents concentration of optically active ions. Survey emission spectra were calibrated in units of the cross section σ_k^{em}(λ) using the Füchtbauer-Ladenburg formula:

$$\sigma_k^{em}(\lambda) = \frac{3\beta\lambda^4 I_k(\lambda)}{8\pi n_k^2 c \tau_{rad} \int [I_\sigma(\lambda) + I_\pi(\lambda)] d\lambda} \quad (1)$$

where I(λ) represents the experimental emission intensity at the wavelength λ, c is the light velocity, n, β and τ_{rad} are the refractive index, branching ratio of emission and radiative lifetime, respectively. The sign k relates to the σ and π parameters. In the case of uniaxial crystal n_k means either n_σ = n_o and n_π = n_e. Emission cross

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