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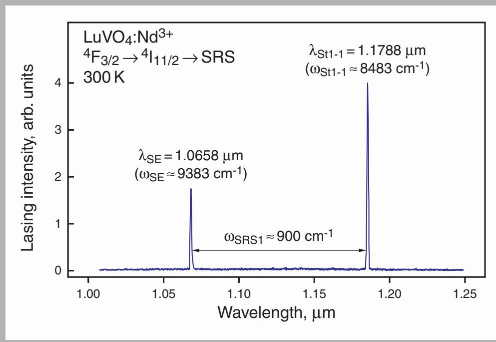
Abstract: Passively Q-switched laser-diode pumped nanosecond Nd^{3+}:$LuVO_4$ self-Raman laser operating at cascade down-converted frequency of the stimulated-emission deriving from the main generation $^4F_{3/2} \rightarrow {}^4I_{11/2}$ channel of neodymium ions has been demonstrated.



The spectra of the self-Raman $LuVO_4$:Nd^{3+} laser (the generation at fundamental wavelength at $\lambda_{SE} = 1.0658$ μm was attenuated by a filter F)

Nanosecond Nd^{3+}:$LuVO_4$ self-Raman laser

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1. Introduction

The discovery of stimulated-Raman scattering (SRS) phenomena in tetragonal YVO_4 and $GdVO_4$ vanadates [1], which have long been known as excellent host-crystals for trivalent lanthanide (Ln^{3+}) lasants ([2], see also [3]) and birefringent optical materials [4], has offered considerable possibilities for the creation, on their basis, of new types of crystalline Raman laser frequency converters and self-Raman (or self-SRS) lasers (see, e.g. [5]), as well as intracavity frequency doubled first-Stokes generation of self-Raman lasers [6]. Quite recently the list of tetragonal SRS-active crystals has been enriched with $LuVO_4$ [7] and $YbVO_4$ [8]. For the last five years a great interest towards Raman lasers, developed on the basis of these vanadates, has been expressed by many groups of researchers (see, e.g. [9–17]). It is essential to note that among the reported developments several self-Raman lasers appeared (see Table 1), which are characterized by peculiar functional simplicity. We must add here also that the Nd^{3+}- and Yb^{3+}-ion doped vanadates YVO_4, $GdVO_4$, and $LuVO_4$ have been successfully used in usual laser-diode (LD) pumped lasers, which can efficient emit in different practical generation regimes (see, e.g. [23]).

In this letter, we report the results on the first performance of a LD-pumped $LuVO_4$:Nd^{3+} self-Raman laser with passively Q-switched nanosecond operation at cascade down-converted frequency of the stimulated-

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Crystal	ω_{SRS} [a)], cm^{-1}	Ln^{3+} lasant, inter-manifold generation channel, SE and first Stokes wavelengths					
		Nd^{3+}				Yb^{3+}	
		$^4F_{3/2} \rightarrow {}^4I_{11/2}$		$^4F_{3/2} \rightarrow {}^4I_{13/2}$		$^2F_{5/2} \rightarrow {}^2F_{7/2}$	
		λ_{SE}, μm	λ_{St1}, μm	λ_{SE}, μm	λ_{St1}, μm	λ_{SE}, μm	λ_{St1}, μm
YVO_4	≈ 890 [1]	1.0641	1.1754 [9,18]	≈ 1.342	≈ 1.525 [19]	≈ 1.018	≈ 1.1195 [11] [b)]
$GdVO_4$	≈ 882 [1]	1.0644 [c)]	1.1744 [10]	≈ 1.341	≈ 1.521 [20]	–	–
	≈ 883 [19] [d)]	1.0651 [c)]	1.1756 [21]				
$LuVO_4$	≈ 900 [7]	1.0658	1.1788	–	–	–	–

[a)] ω_{SRS} is the SRS-promoting vibration mode of crystal.
[b)] According to [11], spectral composition of CW generation depends on output coupler transmission (OC_T) of laser resonator, for given data OC_T = 3.5%. For OC_T = 5.5%, λ_{SE} = 1.014 μm and λ_{St1} = 1.1195 μm.
[c)] These SE wavelengths of [10,21] have some discrepancy with room-temperature Stark level energies of $GdVO_4$:Nd^{3+} crystal from [22].
[d)] In [19] were recorded two additional first Stokes generation at 1.1652 and 1.0950 μm wavelengths, which relate to other SRS-promoting modes of the crystal with energy of ≈ 807 and ≈ 256 cm^{-1}.

Table 1 Crystalline self-Raman lasers on the base tetragonal Ln^{3+}:REVO$_4$ vanadates (Ln^{3+} = Nd^{3+}, Yb^{3+} and RE = Y, Gd, Lu)

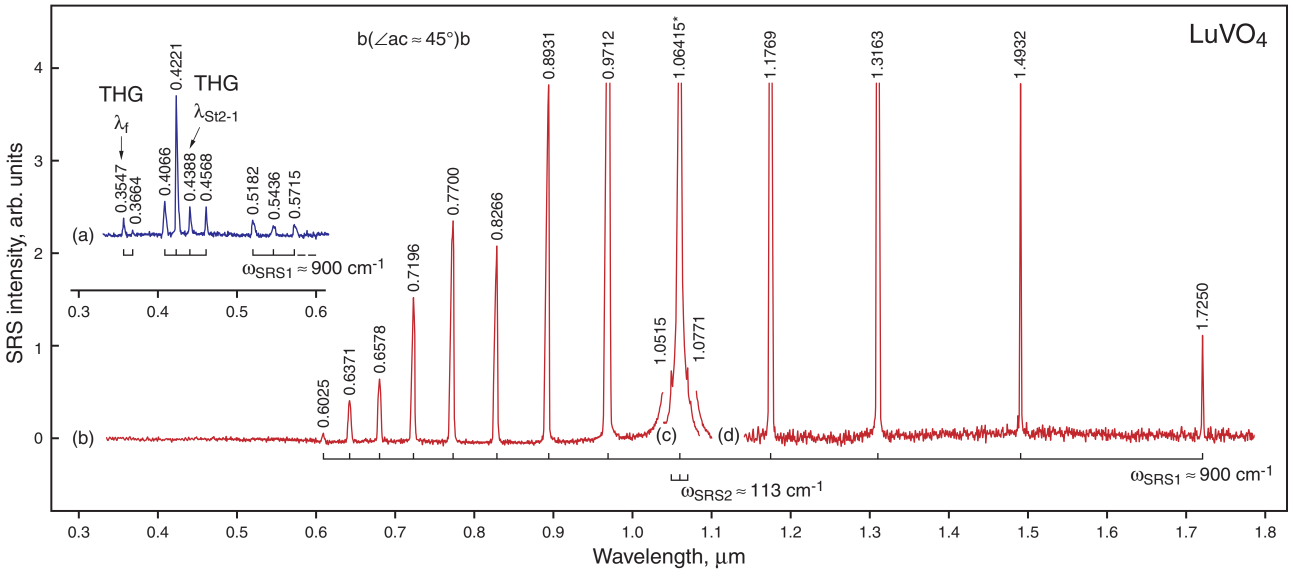


Figure 1 (online color at www.lphys.org) Room-temperature Raman induced $\chi^{(3)}$-lasing spectra of the tetragonal undoped $LuVO_4$ single crystal recorded in excitation geometry $b(\angle ac \approx 45°)b$ with picosecond pumping at 1.06415 μm wavelength of Nd^{3+}:$Y_3Al_5O_{12}$ laser (see details in [7]). The wavelength of all lines (pump line is asterisked) are given in μm, their spectral intensities are shown without correction for the spectral sensitivity of the used analyzing system with Hamamatsu CCD sensors. The spectra (a) – (c) were recorded with a Si-CCD and spectrum (d) with an InGaAs-CCD array sensor. The spacing of the Stokes and anti-Stokes sesqui-octave lasing comb (extended of ≈ 13500 cm^{-1}) components is multiple of the single-SRS-promoting vibration mode with ω_{SRS1} ≈ 900 cm^{-1} of the crystal studied and is indicated by the horizontal scale brackets

emission (SE) deriving from the main generation $^4F_{3/2} \rightarrow {}^4I_{11/2}$ channel of neodymium ions. The research started from our recent discovery of efficient SRS activity in this tetragonal vanadate [7].

2. Crystals and experimental setup

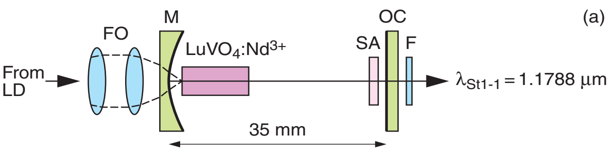
Lutetium vanadate $LuVO_4$, as well as the above-mentioned tetragonal vanadates YVO_4 and $GdVO_4$, have the zircon ($ZrSiO_4$) structure with space group $I4_1/amd$ (point group *4/mmm*). In these materials the Nd^{3+} lasants substitute Lu^{3+} (or Y^{3+}, Gd^{3+}) cations at lattice sites with the D_{2d} symmetry. Several methods for the growth of these crystals (undoped and doped with Ln^{3+} ions) for optical and nonlinear-laser experiments have been described (see Table 2).

The performance of the LD-pumped passively Q-switched self-Raman generation regime in the tetragonal $LuVO_4$:Nd^{3+} vanadate was achieved using a “black gar-

Characteristics [a)]	
Space group	$D_{4h}^{19} - I4_1/amd$ (No. 141)
Unit-cell parameters, Å [24] [b)]	$a = b = 7.026$; $c = 6.234$
Formula units per unit cell	$Z = 4$
X-ray density, g cm^{-3}	$d_x \approx 6.233$
Site symmetry and coordination number (CN) of cations	Lu^{3+}: D_{2d} – CN = 8 [c)]; V^{5+}: D_{2d} – CN = 4
Methods of crystal growth (with Nd^{3+} ions)	Slow cooling in a flux [26], Czochralski [28], floating zone [29]
Hardness (Mohs scale)	≈ 6
Melting temperature, $^\circ$C	≈ 1800
Thermal expansion coefficients, 10^{-6} K^{-1}	$\alpha_{\parallel a-\mathrm{axis}} \approx 2.55$; $\alpha_{\parallel c-\mathrm{axis}} \approx 8.7$
Specific heat, J g^{-1} K^{-1}	≈ 0.34 (at 50°C) [25]
Thermal conductivity, W m^{-1} K^{-1}	$\kappa_{\parallel a-\mathrm{axis}} \approx 7.9$; $\kappa_{\parallel c-\mathrm{axis}} \approx 9.6$ [30] [d)]
Linear optical classification	Positive ($n_o < n_e$) uniaxal
Optical transparency range, μm [e)]	≈ 0.3 – ≈ 5.5
Nonlinearity	$\chi^{(3)}$
Nonlinear coefficient, 10^{-15} cm^2 W^{-1} [f)]	$n_{2(E\parallel c-\mathrm{axis})} = 1.08$; $n_{2(E\perp c-\mathrm{axis})} = 1.34$
Manifestation of nonlinear-laser effects [g)]	SRS, THG [h)], self-SFG(SRS) [h)], Stokes and anti-Stokes frequency comb (see Fig. 1)
Energy of SRS-promoting vibration modes, cm^{-1}	$\omega_{SRS1} \approx 900$; $\omega_{SRS2} \approx 113$
Linewidth (FWHM) of the Raman shifted lines in spontaneous Raman scattering spectra related to SRS-promoting vibration transitions, cm^{-1}	$\Delta\nu_{R1} \approx 5$; $\Delta\nu_{R2} \approx 3.5$
Curent Ln^{3+} lasants [3]	Nd^{3+}, Tm^{3+}, and Yb^{3+}
SE wavelength of $^4F_{3/2} \rightarrow {}^4I_{11/2}$ channel, μm	λ_{SE} =1.0658
Self-Raman lasing wavelength, μm	$\lambda_{St1-1} = 1.1788$
Lifetime of the metastable state $^4F_{3/2}$, μs [i)]	$\tau_{\mathrm{rad}} \approx 82$
Spectroscopic quality parameter [j)]	X_{Nd} ($^4F_{3/2}$) ≈ 1.13

a) The majority of the given data correspond to undoped $LuVO_4$ single crystals.
b) According to [25] $a = b = 7.0389$ Å and $c = 6.2350$ Å, as well as to [26] $a = b = 7.02342$ Å and $c = 6.2316$ Å. For $LuVO_4$:Nd^{3+} ($C_{Nd} \approx 1\%$): $a = b = 7.028(1)$ Å and $c = 6.251(1)$ Å [27].
c) The site for Ln^{3+} activator ions.
d) According to [25] for $LuVO_4$:Yb^{3+} ($C_{Yb} \approx 1.56\%$) $\kappa_{\parallel a-\mathrm{axis}} \approx 5.14$ and $\kappa_{\parallel c-\mathrm{axis}} \approx 5.78$.
e) For ≈ 1-mm thick c-cut plate.
f) Measured by Z-scan method for the ≈ 2-mm thick crystal sample [31].
g) THG, third harmonic generation; self-SFG(SRS), i.e. cascaded self-sum-frequency generation of the arising SRS lasing components at Stokes or anti-Stokes wavelengths and pumping radiation.
h) Non-phase matched.
i) Measured in [26] at low concentration of Nd^{3+} ions ($C_{Nd} \approx 0.04$ at.%).
j) A conservative estimate of the X_{Nd} ($^4F_{3/2}$) $= \Omega_4/\Omega_6$ was made on the base of the known relation $X_{Nd}(^4F_{3/2}) = \Omega_4/\Omega_6$ [32] and the values of the Judd-Ofelt intensity parameters Ω_t reported in [26]. According to [33] for the crystal under investigation $X_{Nd}(^4F_{3/2}) \approx 1.04$.

Table 2 Selected physical properties of tetragonal nonlinear-laser LuVO:Nd^{3+} crystals



Reflection (R) and transmission (T), % (b)

Wavelength	R_M	T_M	R_{OC}	T_{OC}
$\lambda_{SE} = 1.0658$ μm	≈ 99.8	*	≈ 99.8	*
$\lambda_{St1-1} = 1.1788$ μm	≈ 99.8	*	≈ 65	≈ 35
$\lambda_p = 0.808$ μm	≈ 15	≈ 85	**	**

* Minor.
** Lasing crystal absorb ≈ 100% of unpolarized pump radiation.

Figure 2 (online color at www.lphys.org) (a) – schematic of the experimental setup of LD pumped passively Q-switched self-Raman Nd^{3+}:$LuVO_4$ laser; (b) – optical reflection and transmission of cavity components at working lasing wavelengths (see also text)

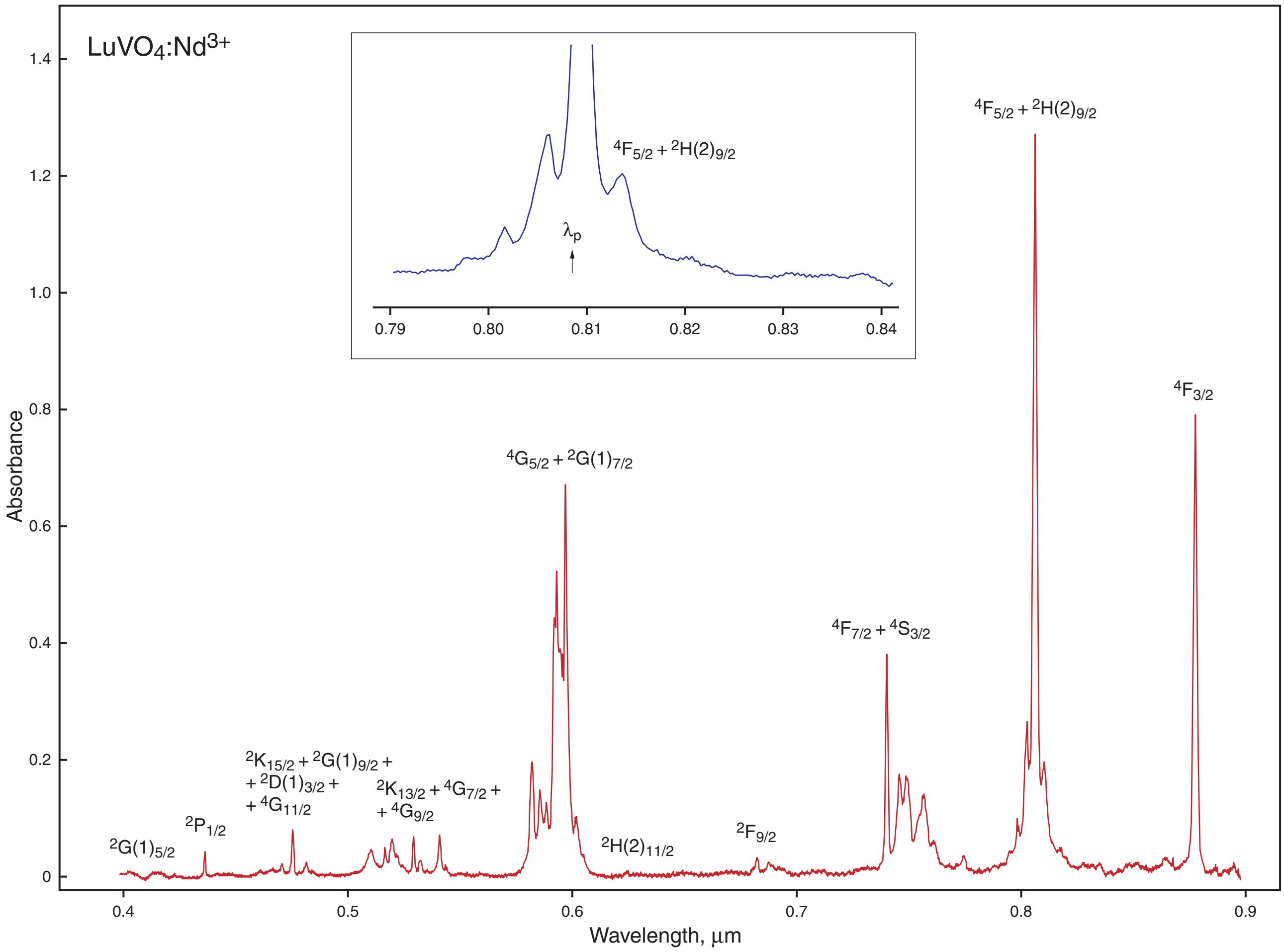


Figure 3 (online color at www.lphys.org) The fragment of room-temperature π-polarized absorption spectrum of $LuVO_4:Nd^{3+}$ crystal ($C_{Nd} \approx 0.45\times10^{20}$ cm^{-3} and $\approx$3-mm thick sample [26]) in the spectral range from 0.4 to 0.9 μm wavelength with the identification of its inter-manifold $^4I_{9/2} \rightarrow {}^{2S+1}L_{J'}$ band-areas. In the frame detail is shown of one part of this spectrum, which is correspond to active pumping channel $^4I_{9/2} \rightarrow {}^2F_{5/2}+{}^2H(2)_{9/2}$

net” ($Y_3Al_5O_{12}$ crystal co-doped with Cr_2O_3 and CaO) as a saturable absorber (SA) and the usual compact laser design. As shown in Fig. 2a, its 35-mm long cavity was formed with a concave (with 4-cm curvature) input mirror (M) and a flat output coupler (OC) having the required dichromic coatings for all “working” wavelengths (see Fig. 1b). The gain medium is provided by a $Nd^{3+}:LuVO_4$ ($C_{Nd} \approx 0.2$ at.%, cut along the c-axis) bar (with a size of $7\times4\times4$ mm^3) with a wide-band antireflection coating of its two plan-parallel active ends. Plane-parallel surfaces of crystalline SA in the form of 2-mm thick plate (with cross-section 10×10 mm^2) were also antireflection coated. The initial transmission of a SA at $\approx$1.1 μm wavelength was about 90%. The pump source was a CW fiber-coupled LD (LIMO GmbH) with a core diameter of $\approx$100 μm in radius and a maximum output power of 5 W. Its 0.808-μm radiation through two-lens focusing optics (FO) with 2-cm focal length and high coupling efficiency was directed into the vanadate bar, which was placed very near the input mirror. As seen from Fig. 3, in our experiment the LD wavelength is not matched with the maximum of absorption peak of the inter-manifold absorption channel $^4I_{9/2} \rightarrow {}^4F_{5/2} + {}^2H(2)_{9/2}$ of Nd^{3+} ions in $LuVO_4$ studied. The lasing crystalline bar was wrapped in an In-foil and mounted tightly in a water-cooled Cu-holder, which was kept a stable temperature of 20°C.

3. Cascaded $^4F_{3/2} \rightarrow {}^4I_{11/2} \rightarrow$ SRS generation

The spectral composition, average output power, and pulse temporal behavior of the Raman induced first Stokes generation of the $LuVO_4:Nd^{3+}$ vanadate were measured by universally accepted methods using a grating spectra analyzer (AQ-6315A), a power meter (Molectron-PM3), and a fast InGaAs PIN photodiode together with a filter (F)

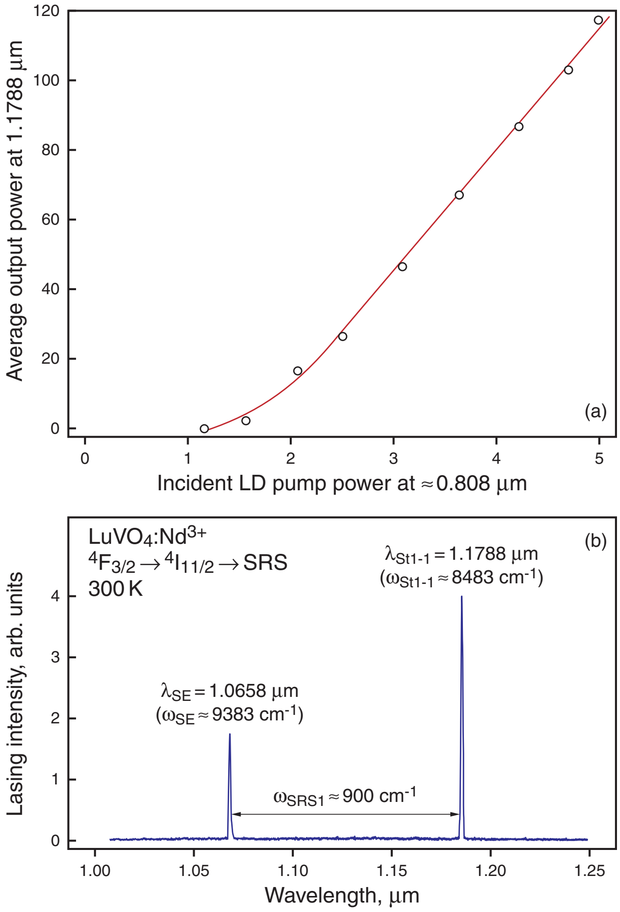


Figure 4 (online color at www.lphys.org) (a) – average output power at cascaded $^4F_{3/2} \rightarrow {}^4I_{11/2} \rightarrow$ SRS generation at $\lambda_{St1-1} = 1.1788$ μm wavelength with respect to incident LD pump power at $\lambda_p \approx 0.808$ μm for pulse repetition rates of ≈ 18 kHz. (b) shows the spectra of the self-Raman $LuVO_4$:Nd^{3+} laser (the generation at fundamental wavelength at $\lambda_{SE} = 1.0658$ μm was attenuated by a filter F)

and a wideband digital Tektronix oscilloscope, respectively. Some of the obtained results are shown in Fig. 4. In the above-mentioned experimental conditions, the "threshold" pump power of the confidently measurable cascaded ($^4F_{3/2} \rightarrow {}^4I_{11/2} \rightarrow$ SRS) self-Raman first Stokes lasing signal at $\lambda_{St1-1} = 1.1788$ μm wavelength was found to be about 1.1 W. With increasing pump power, up to the maximum possible value, the average output power reached ≈ 120 mW; as a consequence, the conversion efficiency is about 5%. Undoubtedly this relatively small efficiency should be further increased by optimizing the setup arrangement, pumping conditions, and all parameters relative to the lasing and Q-switched crystals. It must be added to the given data that at the maximum pump level the repetition rate and pulse duration of the Stokes generation were ≈ 18 kHz and ≈ 2 ns, accordingly.

4. Conclusion

Different manifestations of Raman-induced nonlinear-laser $\chi^{(3)}$-effects in tetragonal $LuVO_4$:Nd^{3+} crystal have been recently discovered [7]; among these manifestations, the almost sesqui-octave Stokes and anti-Stokes comb generation, together with our present performance of the self-Raman laser, as well as numerous results on optical, spectroscopic, and SE characterization (see, e.g. [26,33,34]) showed that this tetragonal crystal is of evident interest for modern laser physics and nonlinear optics. In general, as shown by inspection of Table 3, its thermal properties, as well as SE peak cross-sections (σ^p_{SE}) for inter- Stark laser transitions (for $^4F_{3/2} \rightarrow {}^4I_{11/2}$ and $^4F_{3/2} \rightarrow {}^4I_{13/2}$ channels) and absorption peak cross-section (σ^p_{abs}) for pumping wavelength at 0.808 μm ($^4I_{9/2} \rightarrow {}^4F_{5/2} + {}^2H(2)_{9/2}$), are more favorable compared to the parameters relative to the two other YVO_4 and $GdVO_4$ vanadate laser crystals.

Characteristic	YVO_4	$GdVO_4$	$LuVO_4$
Thermal conductivity, W m^{-1} K^{-1}			
$\kappa_{\parallel a-\mathrm{axis}}$	≈ 5	≈ 10	≈ 7.88
$\kappa_{\parallel c-\mathrm{axis}}$	≈ 5.2	≈ 11.4	≈ 9.63
σ^p_{SE}, 10^{-19} cm^2			
$^4F_{3/2} \rightarrow {}^4I_{11/2}$	≈ 8.0	≈ 7.8	≈ 14.6
$^4F_{3/2} \rightarrow {}^4I_{13/2}$	≈ 2.8	≈ 1.8	≈ 4.3
σ^p_{abs}, 10^{-19} cm^2	≈ 5.7	≈ 5.2	≈ 6.9

Table 3 Room-temperature thermal and spectroscopic characteristics of Nd^{3+}-ions doped YVO_4, $GdVO_4$, and $LuVO_4$ single crystals [33,34]

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