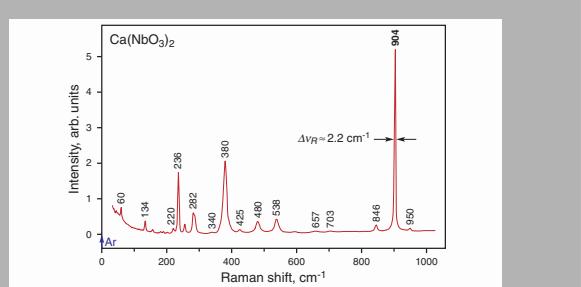


Abstract: A passively Q-switched nanosecond Nd³⁺:Ca(NbO₃)₂ self-Raman laser with 0.808-μm laser-diode pumping has been demonstrated, operating by nonlinear cascaded scheme at converted wavelength of Nd³⁺ one-micron stimulated emission.



The room-temperature first-order spontaneous Raman scattering spectrum (A-modes) of orthorhombic undoped Ca(NbO₃)₂ single crystal

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Q-switched nanosecond Nd³⁺:Ca(NbO₃)₂ crystalline self-Raman laser with single-step cascade SE ($\lambda_{SE} = 1.0615 \mu\text{m}$ of ${}^4\text{F}_{3/2} \rightarrow {}^4\text{I}_{11/2}$ channel) → SRS ($\lambda_{St1} = 1.1741 \mu\text{m}$ of $\omega_{SRS} \approx 904 \text{ cm}^{-1}$ promotion vibration mode) wavelength conversion

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1. Introduction

Investigations in the last decade convincingly evidenced that one of the efficient ways to generate new wavelengths is the development of the so-called self-Raman or self-SRS laser converters (here SRS is the stimulated Raman scattering) on the base of strongly $\chi^{(3)}$ -active insulating

crystals doped with trivalent lanthanide lasants (Ln³⁺), in particular Nd³⁺ ions [1]. The distinctive features of this type of wavelength (frequency) converters are that in these crystals stimulated emission (SE) generation and $\chi^{(3)}$ -nonlinear conversion processes can occur simultaneously, simplified the design of laser devices with very attractive practical potential. Some of them are commercially

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Crystal	Generation of stimulated emission			Stimulated Raman scattering			Ref. ^{a)}
	SE channel	λ_{SE} , μm	Mode ^{b)}	λ_{SRS} , μm	Line ^{c)}	ω_{SRS} , cm^{-1}	
α -KY(WO ₄) ₂	$^4\text{F}_{3/2} \rightarrow ^4\text{I}_{11/2}$	≈ 1.0688	ps	≈ 0.975 ≈ 1.183 ≈ 1.325	ASt ₁ St ₁ St ₂	≈ 905	[4]
α -KGd(WO ₄) ₂	$^4\text{F}_{3/2} \rightarrow ^4\text{I}_{11/2}$	≈ 1.0672	ps, ns ns CW	≈ 0.974 ≈ 1.181 ≈ 1.321 ≈ 1.162 ≈ 1.276 ≈ 1.181	ASt ₁ St ₁ St ₂ St ₁ St ₂ St ₁	≈ 901 ≈ 768 ≈ 901	[2,4,5] [2,6] [7]
				≈ 1.351	ns	≈ 1.538	St ₁ ≈ 901
							[2,8,9]
α -KLu(WO ₄) ₂	$^4\text{F}_{3/2} \rightarrow ^4\text{I}_{11/2}$	1.0702	ns	1.1852	St ₁	≈ 907	[10]
KY(MoO ₄) ₂	$^4\text{F}_{3/2} \rightarrow ^4\text{I}_{11/2}$	1.0669	ns	1.1758 1.1868	St ₁ St ₁	≈ 868 ≈ 947	[10]
NaLa(MoO ₄) ₂	$^4\text{F}_{3/2} \rightarrow ^4\text{I}_{11/2}$	1.0653	ns	≈ 1.177	St ₁	≈ 988	[11]
Ca(NbO₃)₂	$^4\text{F}_{3/2} \rightarrow ^4\text{I}_{11/2}$	1.0615	ns	1.1741	St₁	≈ 904	[this work]
CaMoO ₄	$^4\text{F}_{3/2} \rightarrow ^4\text{I}_{11/2}$	1.0573	ns	1.1656	St ₁	≈ 880	[12]
SrMoO ₄	$^4\text{F}_{3/2} \rightarrow ^4\text{I}_{11/2}$	1.0577	ns	1.1671	St ₁	≈ 980	[12]
SrWO ₄	$^4\text{F}_{3/2} \rightarrow ^4\text{I}_{11/2}$	1.0699	ns	1.1561	St ₁	≈ 922	[12,13]
YVO ₄	$^4\text{F}_{3/2} \rightarrow ^4\text{I}_{11/2}$	1.0641	ns	1.1754	St ₁	≈ 890 ^{d)}	[14]
	$^4\text{F}_{3/2} \rightarrow ^4\text{I}_{13/2}$	≈ 1.342	ns	≈ 1.525	St ₁		[15]
β -LaBGeO ₅	$^4\text{F}_{3/2} \rightarrow ^4\text{I}_{11/2}$	1.0482	ns	0.8972 0.9668 1.1446	ASt ₂ ASt ₁ St ₁	≈ 803	[10]
GdVO ₄	$^4\text{F}_{3/2} \rightarrow ^4\text{I}_{11/2}$	1.0633 ^{e)}	ns	1.1733	St ₁	≈ 882 ^{d)}	[16]
	$^4\text{F}_{3/2} \rightarrow ^4\text{I}_{13/2}$	≈ 1.341	ns, ps	≈ 1.521	St ₁		[17]
LuVO ₄	$^4\text{F}_{3/2} \rightarrow ^4\text{I}_{11/2}$	1.0658	ns	1.1788	St ₁	≈ 900 ^{f)}	[18]
BaWO ₄	$^4\text{F}_{3/2} \rightarrow ^4\text{I}_{11/2}$	≈ 1.055	ns	≈ 1.169	St ₁	≈ 926	[19]
PbMoO ₄	$^4\text{F}_{3/2} \rightarrow ^4\text{I}_{11/2}$	1.0594	ns	1.1668	St ₁	≈ 869	[20]
PbWO ₄	$^4\text{F}_{3/2} \rightarrow ^4\text{I}_{11/2}$	1.0580	ns	1.1695	St ₁	≈ 901 ^{g)}	[10,20,21]

^{a)} Were used articles only in refereed journals.

^{b)} Nonlinear cascade lasing under nanosecond (ns) and picosecond (ps) pulse emission, and continuous-wave (CW) generation.

^{c)} Stokes (St) and ant-Stokes (ASt) nonlinear lasing.

^{d)} SRS-promoting mode was established in [22].

^{e)} SE wavelengths given in [23] have some discrepancy with the room-temperature Stark level energies of GdVO₄:Nd³⁺ crystal (see note (c) for Table 1 in [18]).

The author of [23] measured SE at fundamental wavelength 1.0651 μm and SRS lasing at three wavelengths 1.1756, 1.1652, and 1.0950 μm , which related to different SRS promoting vibration mode of GdVO₄ crystal.

^{f)} SRS-promoting mode was established in [24].

^{g)} SRS-promoting mode was established in [25].

Table 1 Selected self-Raman lasers based on crystals doped with Nd³⁺ ions

available now (see note in [2]). The self-Raman crystalline lasers offer certain advantages (for example, compactness and functional simplicity) compared to usual solid-state Raman lasers in which two separate media – lasing crystal (or glass) and $\chi^{(3)}$ -converting crystal are used. The list of known “neodymium” crystalline self-Raman lasers operated at room-temperature with different pumping sources and mode generation is reported in Table 1. Presently, nu-

merous developments of the last-mentioned type of Raman lasers rank among the scientific and applied aims of many groups of researchers (see, e.g. [3]). The results of the productive investigations mentioned above indicate conclusively that the search of new SRS-active Ln³⁺-ion doped crystals is currently a topical problem because it gives birth to laser sources at new wavelengths.

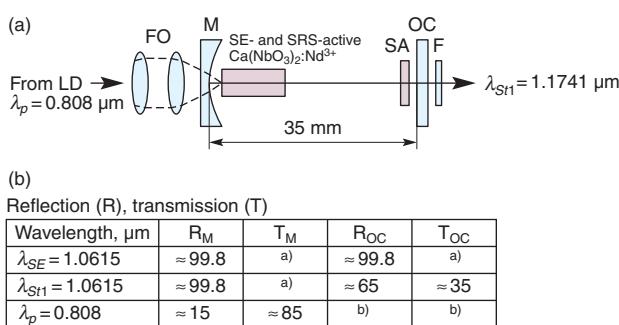


Figure 1 (online color at www.lphys.org) (a) - schematic diagram of LD pumped passively Q-switched Nd³⁺:Ca(NbO₃)₂ self Raman laser; (b) – optical reflection and transmission of cavity components at pumping and lasing wavelengths (see also text)

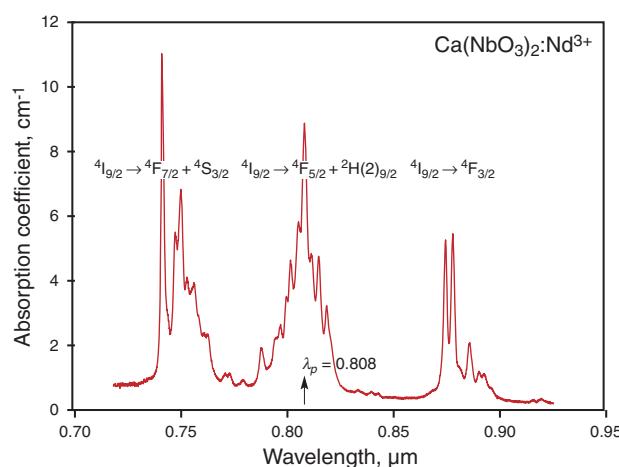


Figure 2 (online color at www.lphys.org) The fragment of room-temperature absorption spectrum of the orthorhombic Ca(NbO₃)₂:Nd³⁺ crystal ($C_{Nd} \approx 2$ at.%, c -cut ≈ 3 -mm plate) in the spectral range from ≈ 0.72 to ≈ 0.93 μm with the identification of its inter-manifold $4\text{I}_{9/2} \rightarrow 2\text{S}^{+1}\text{L}_{J'}$ band-areas, including the pumping channel $4\text{I}_{9/2} \rightarrow 4\text{F}_{5/2} + 2\text{H}(2)_{9/2}$ (see also text)

In this letter, we present results on the first performance of a laser-diode (LD) pumped self-Raman laser on the base of the $\chi^{(3)}$ -active Nd³⁺:Ca(NbO₃)₂ crystal with passively Q-switched nanosecond room-temperature operation at single-step cascade conversion of SE wavelength deriving from the main generation $4\text{F}_{3/2} \rightarrow 4\text{I}_{11/2}$ channel of Nd³⁺ lasants. The research dates back to our recent study of efficient SRS activity in this undoped orthorhombic calcium niobate [26] and our old comprehensive spectroscopic and SE investigations of Ca(NbO₃)₂:Nd³⁺ single crystals [27]. It should be emphasized here that Ca(NbO₃)₂ was the seventh host-crystals for Ln³⁺ lasants [28].

Property	
Space group	$D_{2h}^{14} - P\bar{c}an (Pbcn)$, No. 60
Unit cell parameters, Å [29] ^{a,b)}	$a = 5.757$; $b = 14.97$; $c = 5.225$
Number of formula per unit cell	$Z = 4$
Density, g/cm ³ ^{a)}	≈ 4.77
Melting temperature, °C	≈ 1560
Thermal conductivity, W/m/K ^{c)} [30]	$\kappa_a \approx 6.08$; $\kappa_b \approx 5.71$; $\kappa_c \approx 8.24$
Optical transparency range, μm ^{d)}	≈ 0.3 – ≈ 5.5
Hardness (Mohs scale)	4.5 – 5.5
SE effective peak cross-section, 10^{-19} cm ²	$\sigma_e^p \approx 1.2$ ^{e)}
SRS-promoting vibration mode, cm ⁻¹	$\omega_{SRS} \approx 904$ ^{f)}
First Stokes steady-state Raman gain coefficient, cm/GW	$g_{ssR}^{St1} \approx 2.8$ ^{g)}

^{a)} For undoped crystal.

^{b)} According to a recent refinement within $P\bar{c}an$ setting [31]: $c = 5.22202$; $a = 14.96976$; $b = 5.74724$ Å.

^{c)} For Ca(NbO₃)₂:Nd³⁺ single crystal ($C_{Nd} = 0.894 \times 10^{20}$ cm⁻³).

^{d)} For ≈ 1-mm-thick plate.

^{e)} For $4\text{F}_{3/2} \rightarrow 4\text{I}_{11/2}$ luminescence transition at 1.0615 μm wavelength related to SE generation line.

^{f)} Related to stretching and bending A_g -modes of NbO₆ distorted octahedral (Fig. 3).

^{g)} For the first Stokes SRS line at 1.1741 μm wavelength.

Table 2 Some known room-temperature physical properties of the orthorhombic Ca(NbO₃)₂ and Nd³⁺:Ca(NbO₃)₂ single crystals

2. Crystals and experimental setup

Both undoped and neodymium activated calcium niobate crystals were grown by the usual Czochralski technique. Good optical quality Ca(NbO₃)₂ crystals doped with ≈ 2% Nd³⁺ lasants by weight in the melt were obtained with the stoichiometric amounts of Ti⁴⁺ charge compensators. All calcium niobates were pulled (rate ≈ 6 mm/h and rotation ≈ 30 min⁻¹) in air using an Ir crucible. These crystals were used to fabricate polished samples for nonlinear-laser and spectroscopic measurements.

The performance of Q-switched nanosecond generation regime in self-Raman laser based on the orthorhombic Ca(NbO₃)₂:Nd³⁺ ($C_{Nd} \approx 2$ at.% with charge Ti⁴⁺-compensators) niobate was carried out using a “black garnet” (Y₃Al₅O₁₂ crystal co-doped with Cr₂O₃ and CaO) as a saturable absorber (SA) and widely used simple scheme for compact laser design with laser-diode (LD) pumping. It was practically the same as for LuVO₄:Nd³⁺ self-Raman laser recently described in [18]. As shown in Fig. 1, it is composed of a 35-mm long laser cavity with a 40-mm cur-

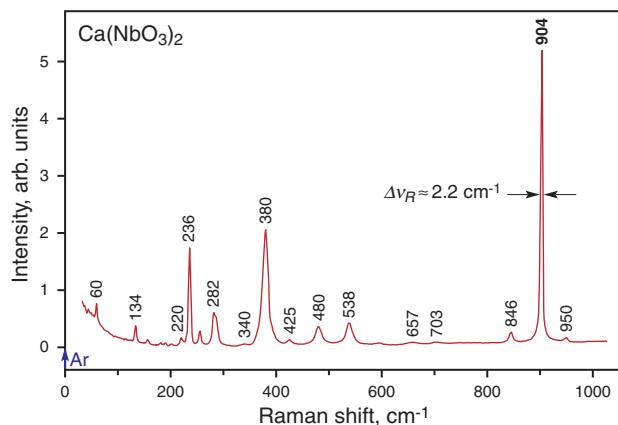


Figure 3 (online color at www.lphys.org) The room-temperature first-order spontaneous Raman scattering spectrum (A-modes) of orthorhombic undoped $\text{Ca}(\text{NbO}_3)_2$ single crystal recorded in back scattering geometry $\approx c$ (bb) $\approx c$ (notation is used by analogy to [32]) under Ar-ion laser $0.488 \mu\text{m}$ wavelength excitation (indicated by arrow). Raman shift

vature concave “pump” mirror (M) and a flat output coupler (OC) having the required dichroic dielectric coatings. The crystalline active $\text{Ca}(\text{NbO}_3)_2:\text{Nd}^{3+}$ element (for SE and SRS) in the form of a rod oriented along the c -axis ($l=5 \text{ mm}$ with $\varnothing=3 \text{ mm}$) with wide-band antireflection coatings of its plane-parallel ends was positioned near the “pump” mirror. On the other side of the compact cavity near its OC a commercial 2-mm thick antireflection coated SA-plate (with cross-section $10 \times 10 \text{ mm}^2$) was placed. The initial transmission of the SA at $\approx 1.1 \mu\text{m}$ wavelength was about 90%. The pump source was a CW fiber-coupled LD (LIMO GmbH) with a core diameter of $100 \mu\text{m}$ and a maximum output power of 5 W at $0.808 \mu\text{m}$ wavelength. Its radiation was directed through two-lens focusing optics (FO) with 20-mm focal length and high coupling efficiency into the lasing $\text{Ca}(\text{NbO}_3)_2:\text{Nd}^{3+}$ crystal. As shown in Fig. 2, the LD wavelength is not matched with the maximum of absorption peak of pump region of Nd^{3+} lasants in the title calcium niobate. The lasing crystalline rod was wrapped in an In foil and mounted tightly in a water-cooled Cu holder, which was kept at a stable temperature of 20°C . Some crystallographic and physical properties of the niobates studied are given in Table 2.

3. Nonlinear laser ${}^4\text{F}_{3/2} \rightarrow {}^4\text{I}_{11/2} \rightarrow \text{SRS}$ cascading

The spectral composition, pulse temporal behaviour of cascaded generation and its average output power of $\text{Nd}^{3+}:\text{Ca}(\text{NbO}_3)_2$ self-Raman laser were measured by universally accepted methods using a spectral analyzer AQ-type, a fast InGaAs PIN photodiode together with a narrowband filter (F) for the first Stokes emission and

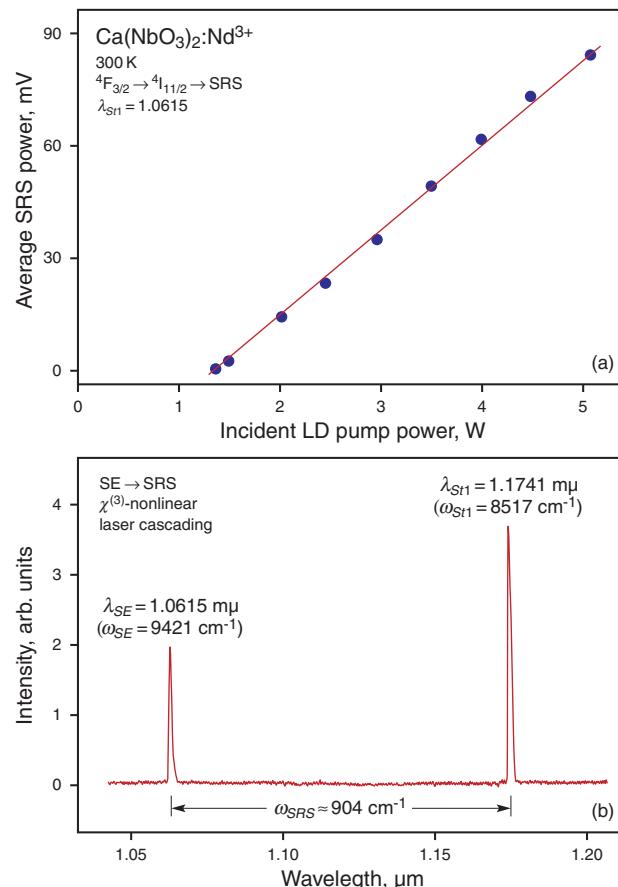


Figure 4 (online color at www.lphys.org) Generation characteristics of nanosecond $\text{Nd}^{3+}:\text{Ca}(\text{NbO}_3)_2$ self-Raman laser: (a) – the dependence of the average output power at $\lambda_{St1}=1.1741 \mu\text{m}$ wavelength as a function of incident LD-pump power at $\lambda_p=0.808 \mu\text{m}$; (b) – the lasing spectrum with SE and SRS lines

a wideband digital Tektronix oscilloscope, as well as a Molelectron-PM3 power meter. Some of obtained results are given in Fig. 4. As seen, the “threshold” pump power of cascade (${}^4\text{F}_{3/2} \rightarrow {}^4\text{I}_{11/2} \rightarrow \text{SRS}$) lasing at $\lambda_{St1}=1.1741 \mu\text{m}$ wavelength was found to be 1.3 W. With increasing pump power up to 5 W, the average output power of nanosecond (with 2.5-ns pulse duration at $\approx 15\text{-kHz}$ repetition rate) nonlinear-laser generation reached $\approx 85 \text{ mW}$ with absolute conversion efficiency of about 2%.

4. Conclusion

As is shown in our research, the Nd^{3+} -ion doped niobates are also attractive crystals for self-SRS conversion processes. We believe that the achieved modest efficiency of one-micron nanosecond cascaded nonlinear-laser generation in the $\text{Nd}^{3+}:\text{Ca}(\text{NbO}_3)_2$ self-Raman laser can be increased by optimizing the spectral pumping condition,

setup arrangements, and the enhancement of the optical quality of title crystal.

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