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Lasing properties of selectively pumped Raman-active Nd^{3+}-doped molybdate and tungstate crystals

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Abstract. **The lasing efficiency of Nd^{3+} ions is studied in laser materials capable of self-Raman frequency conversion. The lasing properties of tungstate and molybdate crystals with the scheelite structure ($SrWO_4$, $BaWO_4$, $PbWO_4$, $SrMoO_4$, $PbMoO_4$) activated with neodymium ions are investigated upon longitudinal pumping by a 750-nm alexandrite laser or a 800-nm diode laser. The slope lasing efficiency obtained for a $Nd^{3+}:PbMoO_4$ laser emitting at 1054 nm is 54.3 % for the total lasing efficiency of 46 %, which is the best result for all the crystals with the scheelite structure studied so far. The simultaneous *Q*-switched lasing and self-Raman frequency conversion were demonstrated in neodymium-doped $SrWO_4$, $PbWO_4$, and $BaWO_4$ crystals.**

Keywords: *solid-state lasers, SRS, tungstates, molybdates, self-frequency conversion.*

1. Introduction

The Nd^{3+} ions in a calcium tungstate $CaWO_4$ crystal were the first rare earth ions in which lasing was obtained in 1961 [1] and which are still being extensively used. This is associated with a convenient energy level diagram of these ions and their good spectroscopic parameters required for obtaining laser action. The spectral range of lasing in Nd^{3+} ions in different matrices is 1030 – 1085 nm for the fundamental ${}^4F_{3/2} - {}^4I_{11/2}$ laser transition and 1310 – 11360 nm for the ${}^4F_{3/2} - {}^4I_{13/2}$ transition [2].

One of the methods for expanding the spectral range of lasing in Nd^{3+} ions is the use of the frequency shift of laser radiation in SRS converters [3, 4]. The frequency shift in such crystal converters achieves 1000 cm^{-1}, which provides the wavelength shift by a few hundreds of nanometres even for a single Stokes shift. It is interesting that nonlinear-optical crystals with the cubic nonlinearity and a high SRS gain can be activated with Nd^{3+} ions and serve simultaneously as an active medium and a self-Raman frequency converter (hereafter, self-conversion) [3 – 5]. Unlike schemes with an additional external Raman converter, such self-frequency conversion lasers are highly stable and compact.

Most of the double tungstates of the type of potassium – gadolinium tungstate $Nd:KGd(WO_4)_2$ (Nd : KGW) or potassium – yttrium tungstate $Nd:KY(WO_4)_2$ (Nd : KYW), tungstates of the type of lead tungstate $Nd:PbWO_4$ and vanadates of the type of yttrium vanadate $Nd:YVO_4$ or gadolinium vanadate $Nd:GdVO_4$ can be used as multifunctional self-conversion crystals. The applications of these materials in self-conversion lasers are described in the literature. The first crystals in which lasing in neodymium ions and self-frequency conversion were simultaneously obtained were Nd : KYW and Nd : KGW crystals [6, 7]. In vanadate crystals activated with neodymium ions, efficient lasing and self-conversion were obtained at 1.17 and 1.5 μm upon [8 – 10].

In this paper, we studied lasing in neodymium ions in tungstate and molybdate crystals with the scheelite structure (Table 1) and compared the results with the lasing parameters of other known laser crystals. The aim of the paper was to compare the lasing efficiency of neodymium ions in tungstate and molybdate crystals with the scheelite structure upon selective pumping under similar experimenatl conditions and to demonstrate the development of self-conversion lasers based on these crystals.

2. Materials and experimental methods

2.1 Tungstate crystals

We studied barium, strontium, and lead tungstate crystals with the scheelite structure which, as has been shown earlier, are efficient matrices for nonlinear-optical Raman converters [11]. Therefore, it was interesting to activate these crystals with neodymium ions to prepare polyfunctional active laser materials for the self-Raman conversion of laser radiation. Our preliminary studies of SRS in tungstate crystals have shown that, depending on the anion of a crystal lattice, the cross section for the Raman gain and the Raman conversion efficiency increase from strontium to barium and then to lead [12, 13]. However, it follows from experiments on the synthesis of neodymium-doped barium tungstate crystals that the distribution coefficient for Nd^{3+} ions in a $BaWO_4$ crystal is very small due to a great difference between the radii of $Ba^{2+}$and Nd^{3+} ions.

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Table 1.

Crystal	Stokes shift/cm^{-1}	Wavelength/nm	
		Lasing in Nd^{3+} ions	SRS lasing
Nd : $SrWO_4$	921.5 [12, 13, 17]	1057 [14, 15, 17]	1171 [18]
Nd : $BaWO_4$	926.5 [12, 13, 20, 21]	1055 [14, 15], this paper	1169, this paper
Nd : $PbWO_4$	904.0 [12, 13]	1058 [22]	1170 [23]
Nd : $PbMoO_4$	869.0 [12, 13]	1059 [24, 25, 27]	1167 [24, 25, 27]
Nd : $SrMoO_4$	887.7 [12, 3]	1057 [26, 27]	1168 [27]

Because of this, the atomic concentration of neodymium ions in the crystal is limited by 0.1 % – 0.2 % even for the crystals co-doped additionally with sodium and niobium ions to compensate for the excess charge [14, 15].

The situation is more promising for strontium tungstate crystals where the difference between the ionic radii of Sr^{2+} and Nd^{3+} ions is less significant, which allowed the synthesis of crystals of high optical quality with a high enough (1 % – 2 %) atomic concentration of neodymium ions [15 – 18].

The most promising material is a neodymium-doped lead tungstate crystal because it combines the best SRS properties of tungstates with the possibility of synthesis of crystals with a high enough concentration of neodymium ions (1 %) and very good spectroscopic properties of the Nd^{3+} ion [19]. The absorption spectra of tungstate crystals are presented in Fig. 1. Note that tungstate crystals cut along the optical axis *c* (polarisation of radiation is perpendicular to this optical axis) do not exhibit the polarisation dependence of the absorption and luminescence spectra of Nd^{3+} ions, as confirmed in our experiments.

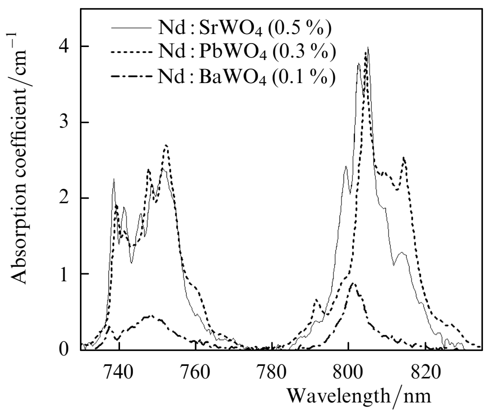

Figure 1. Absorption spectra of tungstate crystals with the scheelite structure for polarisation $E\perp c$.

2.2 Molybdate crystals

We studied strontium and lead molybdate crystals activated with neodymium ions. According to our previous studies, the SRS properties of molybdate crystals surpass those of similar tungstate crystals [12, 28, 29], which is profitably combined in molybdate crystals with good spectroscopic properties of neodymium ions. Investigations have shown that, unlike tungstate crystals, the best spectroscopic properties of neodymium ions in molybdate crystals (the highest absorption and luminescence cross sections) are obtained for radiation polarised parallel to the optical axis *c*.

The absorption spectra of Nd : $SrMoO_4$ and Nd : $PbMoO_4$ crystals for different polarisations of the incident light are presented in Fig. 2. One can see from this figure that the maximum absorption in neodymium-doped molybdate crystals is obtained when the optical axis *c* lies in the crystal plane and the polarisation of the pump radiation is parallel to this axis. For the pump radiation polarised parallel to the optical axis *c* of the crystal, absorption at a wavelength of 750 nm can achieve 3 cm^{-1} for Nd : $SrMoO_4$ and 7 cm^{-1} for Nd : $PbMoO_4$. For the pump radiation polarised perpendicular to the axis *c*, absorption at the pump wavelength 750 nm is less by a factor of 1.5 – 2. Upon pumping at 805 nm and $E\|c$, even higher absorption can be obtained: 5.5 cm^{-1} in a Nd : $SrMoO_4$ crystal and 17 cm^{-1} in a Nd : $PbMoO_4$ crystal.

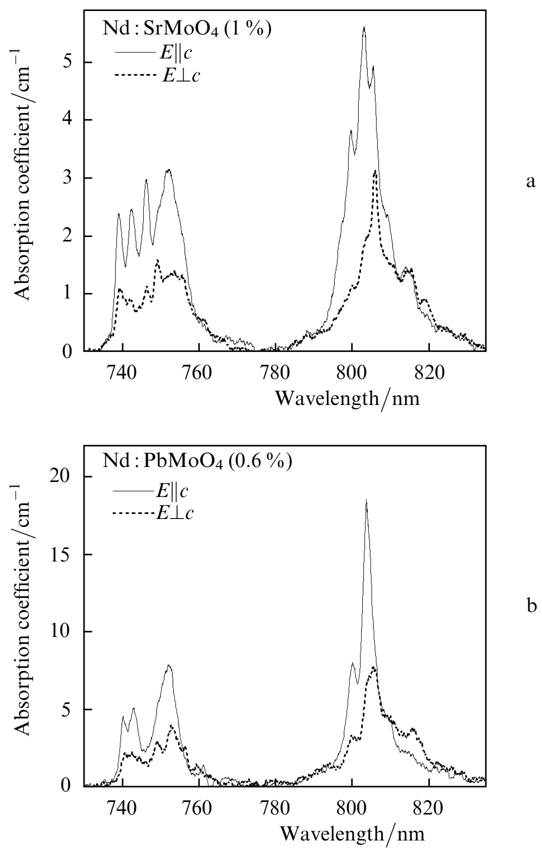

Figure 2. Polarisation absorption spectra of strontium (a) and lead (b) molybdate crystals with the scheelite structure for different polarisations of incident radiation.

2.3 Pumping by an alexandrite laser

In the first part of experiments, pumping was performed by a flashlamp-pumped tunable alexandrite $Cr^{3+}:Al_2O_3:BeO$ laser emitting 50-μs, 300-mJ pulses. This laser conveniently simulated diode pumping at a high enough pulse repetition rate (5 – 10 Hz) with much higher output pulse energy (compared to a diode laser) and was tunable in a rather broad spectral range from 710 to 775 nm. In addition, the divergence of output radiation from an alexandrite laser is low, which allows one to obtain various profiles of the pump beam in a crystal, in particular, with a long waist, which is important upon pumping long crystals with a low concentration of neodymium ions.

The scheme of the optical resonator of neodymium lasers studied in the paper is presented in Fig. 3. The pump radiation was focused into a crystal by a lens with a focal distance of 150 mm to obtain a long enough waist (∼7 mm) of the beam. The optical resonator of the neodymium laser was formed by a dichroic input spherical mirror with the radius of curvature $r = 500$ mm and high transmission at the pump wavelength (740 – 750 nm) and 100 % reflection at the lasing wavelength in the 1-μm region. The length of the optical resonator was 100 mm. Such a large length is caused by the necessity of placing mirrors far enough from the pump beam waist to avoid an optical damage at their surface. The crystals were pumped by tuning the alexandrite laser tuned to the maximum of the absorption spectrum of neodymium ions at 752 nm.

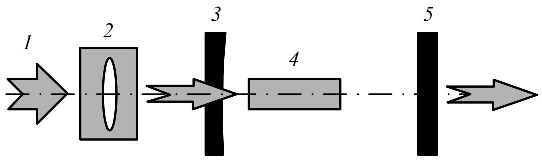

Figure 3. Optical scheme of the resonator of the lasers studied: (*1*) pump laser beam; (*2*) focusing system; (*3*) input dichroic mirror; (*4*) crystal under study; (*5*) output mirror.

2.4 Pumping by a diode laser

In the second part of experiments, pumping was performed by a fibre-coupled diode laser and a special system for beam profiling at the fibre output, which provided a long enough ($l = 4$ mm) waist of the pump beam. The diameter of the pump beam waist was 400 μm. Because the output radiation was coupled out of the diode-laser resonator through a fibre, it had a circular polarisation, and therefore no polarisation dependence of the output lasing parameters of the neodymium-doped crystals pumped by the laser diode was observed in our experiments. The scheme of the optical resonator of the neodymium laser was similar to this shown in Fig. 3, the only difference being that the input dichroic mirror was plane and transparent at 805 nm and spherical mirrors ($r = 120$ mm) with different reflection coefficients (2 % and 8 %) were used as the output mirror. The output emission line of the diode laser was not temperature-tuned to the maximum of the absorption spectrum of neodymium except specially mentioned cases.

3. SRS lasing experiments

To obtain high radiation power densities required to achieve the self-conversion threshold, lasers based on neodymium-doped SRS crystals were *Q*-switched by using a $LiF:F_2^-$ crystal passive *Q* switch. Because lasing of neodymium occurred at the $^4F_{3/2} - ^4I_{11/2}$ transition at ∼1 μm, SRS lasing at the first Stokes component was observed at ∼1.17 μm. SRS lasing was obtained by using a special optical resonator formed by the input spherical mirror with the radius of curvature of 0.5 m having a high transmission at the pump wavelength and a high reflection in the 1000 – 1250-nm region. The output plane mirror partially transmitted light at the wavelength of the first Stokes component and had a high reflection coefficient at the lasing wavelength of Nd^{3+} ions.

The temporal parameters of laser radiation were measured with a fast FNSPE vacuum diode with a 1000-V power supply. The pass band of the diode exceeded 2 GHz. The pulse energy was measured with a Molectron-J25 power meter with a sensitivity of 8.59 V J^{-1} connected with a 500-MHz Tektronix-3052B oscilloscope. The average output power of laser radiation was measured with a Molectron-EMP 2000 energy and power meter. The emission spectrum of the laser was recorded with an Oriel 77250 monochromator (with the 50-μm slit width) coupled with an Electrim EDC-1000HR CCD camera.

4. Results and discussion

4.1 Pumping by an alexandrite laser

Note that, despite the low threshold energy, the lasing efficiency of $Nd:BaWO_4$ was the lowest of the efficiencies of all the crystals studied (the slope efficiency is 12.2 %), which is explained by a very low concentration of neodymium ions in this crystal and by the necessity of using a long crystal ($l = 37.7$ mm). This results in the non-optimal distribution of the pump intensity over the crystal length and a poor matching between the size of the pumped region and the mode of the optical resonator. A much higher lasing efficiency was obtained for a $Nd:SrWO_4$ crystal whose the slope efficiency was 46 % for the output mirror with the reflectivity of 60 %. For the output mirror with the reflectivity of 50 %, the lasing threshold increased by a factor of 2.5.

The lasing thresholds for both lead tungstate crystals with atomic neodymium concentrations 0.3 % and 0.6 % and the reflectivity of the output mirror equal to 60 % were almost the same and approximately twice the threshold energy of $Nd:BaWO_4$ and $Nd:SrWO_4$ crystals, which is probably explained by a lower optical quality of $Nd:PbWO_4$ crystals resulting in higher parasitic losses at the crystal ends and inside it. For the output mirror with the reflectivity of 35 %, the lasing thresholds for the two crystals with different concentrations of neodymium were still very close, but unlike the $Nd:SrWO_4$ crystal, the lasing threshold with the more transparent output mirror increased for both crystals only by a factor of 1.4.

The highest slope lasing efficiency (52.9 %) was obtained for the $Nd:PbWO_4$ laser (0.6 %) with the output mirror reflectivity of 35 %, which is approximately twice the slope lasing efficiency for the $Nd:PbWO_4$ laser with the concentration of neodymium ions equal to 0.3 %. The obtained results demonstrate good lasing properties of Nd^{3+} ions in the $PbWO_4$ crystal. A further improvement of the optical quality of crystals and optimisation of the concentration of neodymium ions in these crystals should further increase the lasing efficiency.

Along with the dependences of the output energy of the $Nd:PbMoO_4$ laser on the pump energy for different reflectivities of the output mirror, we also measured the dependences of the output energy for two orthgonal polarisations of the pump radiation – along the optical axis of the crystal ($E \| c$) and perpendicular to it ($E \perp c$). Due to a weaker absorption of pump radiation, the lasing efficiency was less almost by half in the case of perpendicular polarisation. The best results were obtained for the $Nd:PbMoO_4$ laser with the atomic concentration of neodymium ions of 0.3 % and the output mirror reflectivity of 60 %. The slope lasing efficiency for this laser was 54.3 %. Note that such a high lasing efficiency was obtained at the concentration of neodymium ions smaller by half than that in the $Nd:PbWO_4$ (0.6 %) crystal next in the lasing efficiency.

It should be emphasised that in both cases the best results were obtained for crystals containing lead cations (Pb^{2+}). A high covalence of the lead atom bonding in crystals leads to the highest absorption and luminescence cross sections of neodymium ions, which is extremely important for achieving efficient pumping and high lasing cross section for neodymium ions. In addition, Pb^{2+} ions have a radius close to that of neodymium ions and admit a comparatively high level of activation of crystals with Nd^{3+} ions. At the same time, crystals containing lead ions are also the most efficient, having a low threshold and a high Raman gain, which makes it possible to develop highly efficient self-conversion lasers.

We compared the lasing parameters of neodymium ions in new tungstate and molybdate crystals with parameters obtained under similar conditions for the optimised commercial Nd : KGW crystal with the atomic concentration of neodymium ions 2.2 % and AR coatings deposited on the ends of the active element. The best results on lasing upon pumping by the alexandrite laser are presented in Fig. 4.

4.2 Pumping by a diode laser

Note at once that in all experiments with diode pumping, the wavelength of the diode laser was not tuned to the maximum of the absorption line of neodymium ions. The exception was made for a Nd : KGW crystal which was pumped at a wavelength tuned to the maximum of the absorption band of neodymium ions to obtain the maximum output energy. However, in each experiment we measured the real absorption in the crystal under study at the pump wavelength.

The highest lasing threshold (1.4 mJ) and one of the lowest slope lasing efficiencies (9.2 %) was observed, as in the previous experiment, for the $Nd:BaWO_4$ laser because of the low concentration of neodymium ions and a long length of the crystal ($l = 37.7$ mm).

The lasing threshold in the $Nd:PbWO_4$ crystal with a lower concentration of neodymium ions was almost twice higher and the lasing efficiency was lower by half compared to the crystal with a higher concentration of neodymium ions. The lasing threshold for the $Nd:PbWO_4$ laser with the atomic concentration of neodymium ions of 0.6% and the output mirror reflectivity of 98 % was 0.47 mJ and the slope efficiency was 13.4 % (the total lasing efficiency was 12 %).

Both molybdate crystals ($Nd:SrMoO_4$ and $Nd:PbMoO_4$) have a rather high lasing threshold ($\sim$1 mJ), which is slightly better than that for the barium tungstate crystal. This indicates that the optical quality of these molybdate crystals is rather low. The slope lasing efficiency for $Nd:SrMoO_4$ was 16.4 %, while for $Nd:PbMoO_4$, as upon pumping by the alexandrite laser, the highest slope efficiency equal to 21 % was obtained (the total efficiency was 17 %). Thus, strontium and lead molybdate crystals pumped by nonpolarised radiation from a diode laser have the highest lasing efficiency compared to all other studied crystals with the scheelite structure.

Figure 5 presents the dependences of the output energy of neodymium-doped lead tungstate and molybdate crystal lasers on the absorbed pump energy of the diode laser. The lasing parameters of all the crystals studied are summarised in Table 2 for comparison. The table presents the threshold lasing energy and the maximum slope (η_{slope}) and total (η_{tot}) lasing efficiencies for pumping by the alexandrite and diode lasers. One can see from Table 2 that neodymium-doped tungstate and molybdate crystals with the scheelite structure

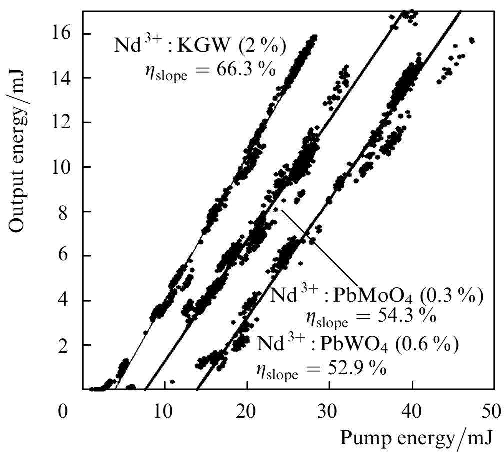

Figure 4. Dependences of the output energy of $Nd^{3+}:PbWO_4$, $Nd^{3+}:PbMoO_4$, and Nd^{3+} : KGW lasers on the alexandrite laser pump energy.

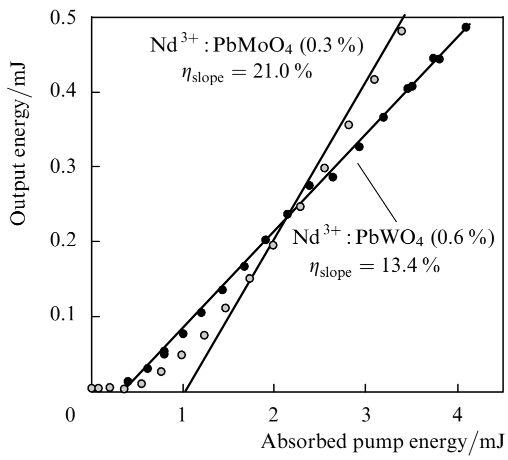

Figure 5. Dependences of the output energy of $Nd^{3+}:PbWO_4$ and $Nd^{3+}:PbMoO_4$ lasers on the absorbed pump energy of the diode laser.

Table 2.

Crystal	Crystal length/mm	Atomic concentration of Nd^{3+} (%)	Absorption (%)	Threshold energy/mJ	η_{slope} (%)	η_{tot} (%)	Polarisation
KGW : Nd	8	2.2	92.1	3.7	32.6	28	⊥
			92.1	4	66.3	52	‖
$PbMoO_4$: Nd	4.9	0.3	43.9	7	21.8	16	⊥
			66.8	7.6	54.3	46.4	‖
$PbWO_4$: Nd	9.7	0.6	97.7	13.9	52.9	33.3	⊥
$SrWO_4$: Nd	45	1	99.5	8.7	48.5	42	⊥
$PbWO_4$: Nd	8.4	0.3	89.6	16.6	25.9	15	⊥
$SrMoO_4$: Nd	1.5	1	38.5	7.5	24.4	17.2	⊥
$BaWO_4$: Nd	37.7	0.1	50.6	6.9	12.3	9.4	⊥
KGW : Nd	8	2.2	88	0.69	44.6	39	⊕
$PbMoO_4$: Nd	4.9	0.3	69	1.04	21.0	17	⊕
$SrMoO_4$: Nd	1.5	1	32	1.05	16.4	12.3	⊕
$PbWO_4$: Nd	9.7	0.6	98	0.47	13.4	12	⊕
$BaWO_4$: Nd	37.7	0.1	80	1.38	9.2	7	⊕
$PbWO_4$: Nd	8.4	0.3	63	0.89	6.3	4.5	⊕

have a high lasing efficiency which is close to that of an optimised laser based on a commercial Nd : KGW crystal, which is widely used in efficient laser systems. We obtained the slope efficiency for the Nd : $PbMoO_4$ scheelite crystal laser equal to 54.3% (the total efficiency was 46 %) upon pumping by the alexandrite laser and to 21 % (the total efficiency was 17 %) upon pumping by the diode laser, which is the best result among all the lasers studied in this paper. Note that tungstate and molybdate crystals with the scheelite structure have a much higher SRS gain and a higher SRS conversion efficiency than that for the known KGW crystal.

4.3 Demonstration of self-Raman frequency conversion

All crystals in which lasing frequency self-conversion is possible were studied in the SRS optical resonator. Self-conversion was observed in Nd : $BaWO_4$ and Nd : $SrWO_4$ crystals pumped by the alexandrite laser and Nd : $PbWO_4$ crystals pumped by the diode laser. The parameters of Raman lasers and obtained results are presented in Table 3.

The maximum pump energy of the alexandrite laser absorbed in the Nd : $BaWO_4$ crystal was 30 mJ. When the output mirror of the optical resonator had partial transmission at the lasing wavelength of Nd^{3+}, simultaneous lasing was observed in the Nd : $BaWO_4$ crystal at 1055 nm and the corresponding first Stokes component at 1169 nm. The laser emitted 4.4-ns, 0.69-mJ pulses at 1055 m and 2.3-ns, 0.25-mJ pulses at 1169 nm. By using the output mirror partially transmitting only at the SRS wavelength ($R_{1055} > 99$ %, $R_{1169} = 60$ %), we obtained self-conversion of radiation with the maximum output pulse energy of 0.8 mJ for the first Stokes component at 1169 nm for the pulse duration of 1.3 ns corresponding to the peak power of 615 kW.

Table 3.

Parameters	Laser crystals		
	Nd : $SrWO_4$	Nd : $BaWO_4$	Nd : $PbWO_4$
Atomic concentration of Nd^{3+} ions (%)	0.5	0.1	0.6
Crystal length/mm	45	37.7	9.7
Pump wavelength/nm	752 (alexandrite laser)	752 (alexandrite laser)	803 (diode laser)
Resonator length/mm	140	140	75
Output mirror reflectivity (%)			
at $\lambda = 1.06$ μm	99.9	99.9	99.5
at $\lambda = 1.17$ μm	47	47	99.5
Initial transmission of a LiF : F_2^- passive Q switch	5	60	60
Threshold lasing energy/mJ	79	14	3.5
Laser wavelength/nm	1170	1169	1158 and 1170
SRS pulse duration/ns	2.9	1.3	4.8
SRS pulse energy/mJ	4.5	0.8	–
SRS laser pulse peak power/kW	1550	620	–

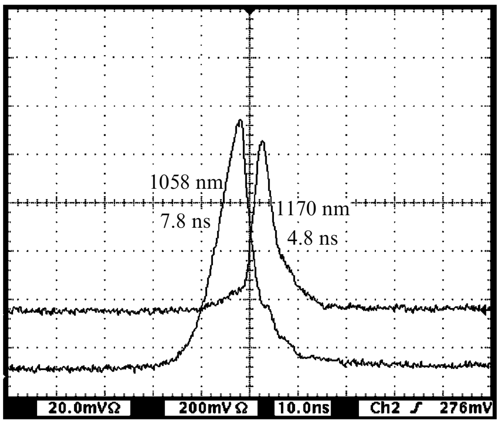

Figure 6. Oscillograms of laser pulses upon the simultaneous Raman conversion of radiation in the Nd : $PbWO_4$ crystal pumped by the diode laser.

The maximum energy of the 1.9-ns self-lasing pulses emitted by the Nd : $SrWO_4$ laser pumped by the alexandrite laser was 4.5 mJ. Figure 6 shows the time dependences of laser pulses at the frequency-shifted first Stokes component for the Nd : $PbWO_4$ crystal pumped by the diode laser. Unfortunately, the optical quality of the Nd : $PbWO_4$ crystal, as mentioned above, was poor and the breakdown of the crystal surface occurred upon *Q*-switching, which prevented the measurement of the self-conversion pulse energy.

One can see from Fig. 6 that due to the cubic nonlinearity of Raman conversion, the output pulse in self-conversion systems considerably shortens compared to the pump pulse, which allows one to obtain nanosecond and subnanosecond pulses in compact laser systems of this type. The conversion efficiency in self-conversion lasers based on tungstate and molybdate crystals can achieve 70 % – 90 %, which allows the generation of frequency-shifted short laser pulses for various applications.

5. Conclusions

We have obtained efficient lasing in neodymium-doped SRS-capable tungstate and molybdate crystals, which is comparable with the lasing efficiency of the well-known and widely used Nd : KGW laser. The tungstate and molybdate crystals with the scheelite structure have a much higher Raman gain and a higher Raman conversion efficiency than the KGW crystal. The slope efficiency of the Nd : $PbMoO_4$ laser is 54.3 % (the total efficiency is 46 %) upon pumping by the alexandrite laser and 21 % (the total efficiency is 17 %) upon pumping by the diode laser. This is the best result for all the crystals with the scheelite structure studied so far.

Self-Raman conversion has been demonstrated in Nd : $SrWO_4$ and Nd : $BaWO_4$ lasers pumped by the alexandrite laser and in the Nd : $PbWO_4$ laser pumped by the diode laser. We have failed to obtain such a conversion in a commercial Nd : KGW laser under similar conditions.

Further improvement of the optical quality of tungstate and molybdate crystals and the optimisation of the concentration of neodymium ions should increase their lasing efficiency at $\sim$1 μm. The self-conversion of laser radiation of neodymium ions in these crystals leads to the generation of considerably shortened output pulses with a high peak power at new wavelengths in the region between 1.2 and 1.5 μm.

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