

# NaLa(WO<sub>4</sub>)<sub>2</sub> and NaY(WO<sub>4</sub>)<sub>2</sub> Raman Converters for Picosecond Pulses

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**Abstract:** Quasi-steady state SRS with 1.7 ps pulses is investigated in tetragonal NaY(WO<sub>4</sub>)<sub>2</sub> and NaLa(WO<sub>4</sub>)<sub>2</sub> crystals in the regimes of collinear and noncollinear SRS-assisted four-wave-mixing. Single-pass conversion efficiency of 31% is reported.

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Picosecond light sources generating pulses with the pulse length in 1 ps - 5 ps range are limited to few laser hosts and rather narrow range of wavelengths. Typically optical parametric oscillators and/or amplifiers are used to access wavelengths different from those of the laser. In fact, stimulated Raman scattering (SRS) could offer certain advantages as wavelength shifting mechanism, primarily due to good temporal coherence, high contrast, high efficiency and simplicity of experimental arrangement. That presupposes that the SRS process is operating in so called steady-state or close to steady state regime, i.e., when the pump pulse length is much larger than the vibrational relaxation time,  $\tau \gg T_R = [\pi\Delta\Omega]^{-1}$ , where  $\Delta\Omega$  is the frequency bandwidth of corresponding spontaneous Raman scattering line. Recently, generation of 5.9 ps mode-locked pulses with self-Raman frequency shifting has been demonstrated in Nd:YVO<sub>4</sub> [1]. Monoclinic and tetragonal tungstate crystals have been investigated as prospect SRS media with high Raman gain [2]. An additional advantage of tungstate crystals is the possibility of doping them with a wide range of rare-earth ions making these materials potentially versatile laser and SRS hosts. Most of the SRS-active and most intense phonon modes in tungstate crystals have relaxation times of the order of 2 ps, mandating the use of rather long, 20 ps - 100 ps, pulses [3, 4] for efficient SRS interaction. Notable exceptions are disordered tetragonal double tungstates such as NaY(WO<sub>4</sub>)<sub>2</sub>, NaLa(WO<sub>4</sub>)<sub>2</sub>, which have subpicosecond relaxation times for the most intense Raman lines [2, 5]. The increased linewidth of Raman scattering, however, comes at a cost of decreased Raman gain. On the other hand, the higher peak intensities afforded by shorter pulses can at least partly compensate for this gain decrease. In this work we investigate picosecond SRS performance in NaY(WO<sub>4</sub>)<sub>2</sub>, NaLa(WO<sub>4</sub>)<sub>2</sub> pumped by the optical pulses with the pulselength of the order of 2 ps, which is the typical pulse length produced by mode-locked picosecond Ti:Sapphire or Yb<sup>3+</sup> - based lasers and amplifiers.

NaLa(WO<sub>4</sub>)<sub>2</sub> and NaY(WO<sub>4</sub>)<sub>2</sub> isostructural single crystals have been grown by the Czochralski method. Details of the crystal growth procedures can be found in Ref. 5. Samples were oriented by Laue X-ray diffraction patterns and later the surfaces polished to laser grade. The surfaces of the samples were left uncoated. Spontaneous Raman scattering measurement in these materials [2, 5], reveal two prominent phonon lines located around  $\Omega_1 = 325 \text{ cm}^{-1}$  and  $\Omega_2 = 914 \text{ cm}^{-1}$  (the exact positions depending on measurement geometry) with relaxation time of the order of 0.8 ps. These lines have been attributed to the  $A_g(\nu_3)$  and  $A_g(\nu_1)$  stretching optical modes of tetrahedral WO<sub>4</sub><sup>2-</sup> ions, respectively [2]. The pump pulses were derived from a Ti:Sapphire regenerative amplifier system operating at 1 kHz repetition rate and generating 1.7 ps pulses. The central wavelength was 822 nm and the FWHM spectral width of the pump pulse was 1.6 nm. The pump was propagating along *a*-crystal axis and the physical length of the samples along this direction was 6.2 mm and 8 mm for NaLa(WO<sub>4</sub>)<sub>2</sub> and NaY(WO<sub>4</sub>)<sub>2</sub>, respectively. The SRS measurements were performed in a(cc)a configuration, which showed the lowest SRS thresholds. The pump beam was loosely focused to 170  $\mu\text{m}$  beam radius inside the crystals. The loose focussing was intentionally chosen in order to avoid broadening of the pump spectrum and white light continuum generation before SRS takes place. The output radiation after the crystal was collected by a 50 mm diameter *f*=50 mm lens and focused into the entrance of a spectrum analyzer (ANDO AQ-6315A). In order to prevent large pump intensities reaching lens and the spectrum analyzer, large part of the pump beam before the lens was blocked with small circular beam-block. Wide-angle collection optics was intentionally chosen in order to be able to measure the SRS radiation possibly containing angular dispersion. This is important in the regime of transient SRS, characterized by a wide-angle scattering and also when the SRS-mediated four-wave-mixing takes place at higher pump powers [6].

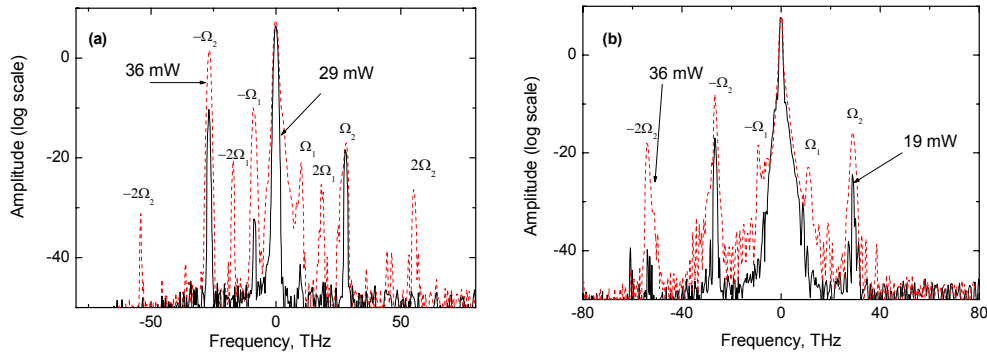


Fig. 1. picosecond SRS spectra in NaLa(WO<sub>4</sub>)<sub>2</sub> (a) and NaY(WO<sub>4</sub>)<sub>2</sub> (b) for the pump powers of 29 mW, 19 mW (solid line in (a) and (b) respectively) and 36 mW (dashed lines).

SRS threshold in NaLa(WO<sub>4</sub>)<sub>2</sub> was reached at 8.9 GW/cm<sup>2</sup>, while in NaY(WO<sub>4</sub>)<sub>2</sub> it was around 8 GW/cm<sup>2</sup>. Considering slightly different lengths of the samples it can be concluded that the SRS gain in both materials is similar of around 4 cm/GW assuming steady-state SRS process. This approximation is, however, not strictly valid considering that the pump pulses are only about 2-times longer than the relaxation time. On the other hand the representative spectra measured in NaLa(WO<sub>4</sub>)<sub>2</sub> and NaY(WO<sub>4</sub>)<sub>2</sub> for two different pump powers and shown in Fig.1 (a) and (b), respectively, reveal well-defined SRS peaks corresponding to the above mentioned

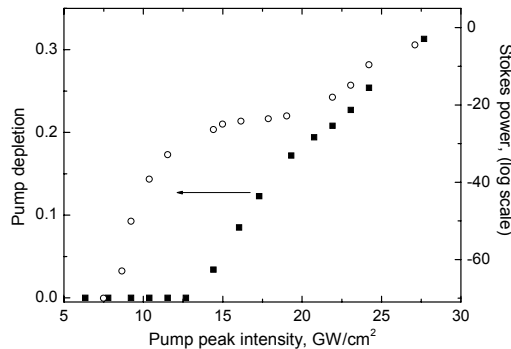


Fig.2 Amplitude of the first Ω<sub>2</sub> Stokes (open circles), pump depletion (solid squares) as a function of pump peak intensity in NaY(WO<sub>4</sub>)<sub>2</sub>

vibrational modes Ω<sub>1</sub> and Ω<sub>2</sub>. The mode Ω<sub>2</sub> is the strongest in both materials producing both the Stokes and anti-Stokes cascades. Moreover, in NaY(WO<sub>4</sub>)<sub>2</sub> the mode Ω<sub>1</sub> was not very pronounced even at higher pump intensities making the SRS spectrum much simpler. Pump depletion and the dependence of the first Ω<sub>2</sub> Stokes amplitude on the pump intensity is shown in Fig.2 for NaY(WO<sub>4</sub>)<sub>2</sub>. Up to the pump intensities of 15 GW/cm<sup>2</sup>, single SRS cascade is active, while at higher intensities SRS cascades develop so the power growth in the first Stokes slows down. Above 20 GW/cm<sup>2</sup> SRS assisted noncollinear four-wave mixing becomes active, generating structured rings of anti-Stokes cascades. Single pass pump depletion up to 31% have been measured.

In conclusion, NaLa(WO<sub>4</sub>)<sub>2</sub> and NaY(WO<sub>4</sub>)<sub>2</sub> crystals with short vibrational relaxation times offer substantial SRS gain for relatively short ~2 ps pulses and can be considered as intracavity laser frequency shifters.

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