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Diode array pumped, non-linear mirror Q-switched and mode-locked Nd : YVO_4 laser – a good tool for powder SHG measurement

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Abstract. A non-linear mirror consisting of a lithium triborate crystal and a dichroic output coupler are used to mode-lock (passively) an Nd : YVO_4 laser, pumped by a diode laser array. The laser can operate both in cw mode-locked and simultaneously Q-switched and mode-locked (QML) regime. The peak power of the laser while operating in QML regime is much higher but pulses suffers from poor amplitude stability. The incorporation of an acousto-optic modulator as an active Q-switch enhances the stability of the QML pulse envelope. The second-order non-linearity of powdered crystalline urea is conclusively measured with respect to KDP while the laser is operating in passively Q-switched and passively mode-locked regime as well as in actively Q-switched and passively mode-locked regime.

Keywords. Non-linear mirror; mode-locking; Q-switching; powder SHG.

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1. Introduction

It is very important for the crystal growers to know the non-linear optical property of their newly synthesized compound before going for its bulk growth. The popular method of being assured of the second-order non-linear optical property is second harmonic generation (SHG) of powdered crystalline sample – a method introduced by Kurtz and Perry in 1968 [1]. Kiguchi *et al* [2] reported measurement of second-order non-linearity of a powdered sample by phase-matched SHG of evanescent wave under total reflection conditions. But this method requires a high index hemispherical prism for total internal reflection from the sample. As such, the use of this method is limited only for low index materials. Most of the experiments reported so far on powder SHG measurement, used nanosecond Q-switched laser

[3,4]. The non-linear interaction being an instantaneous phenomenon, high peak power optical pulse is more favorable than high average power. Moreover, powder SHG being a non-phase-matched second-order phenomenon, high peak power laser is the only option for better SHG efficiency and comprehensive prediction of effective non-linearity of newly synthesized material. A cw mode-locked (ML) picosecond pulse train can be a good choice, but a Q-switched and mode-locked (QML) pulse train is ideal for this measurement. A cw ML laser gives picosecond pulses with high amplitude stability whereas for QML, picosecond mode-locked pulses lie underneath a Q-switched nanosecond pulse envelope. In QML, the peak power is reasonably greater than the other two regimes of operation mentioned above. The other definite advantage of QML lasers for powder SHG measurement is that the non-linearity in generation can be verified by simultaneous recording of fundamental and SH pulse train envelope.

The non-linear mirror mode-locked (NLMML) laser [5,6] considered here, works in the principle of cascaded second-order non-linear process. When two second-order non-linear optical processes occur in a crystal to give an effective third-order non-linearity that is different from the intrinsic third-order non-linearity of the crystalline medium, the phenomenon is called a cascaded second-order process. Recently, a number of applications have been reported for cascaded second-order effects such as all-optical switching [7,8], nearly degenerate four-wave mixing [9,10], gain, transistor action [11], pulse compression, mode-locking of laser [5,6,12] and spatial optical solitons [13]. The large effective $\chi^{(3)}$ due to cascading has been used for mode-locking of cw solid state laser in two schemes: one using the non-linear phase-shift, i.e., equivalent to $\mathrm{Re}[\chi^{(3)}]$, and it is called cascaded second-order non-linear mode-locking (CSM) [14] whereas the other one uses non-linear amplitude modulation, i.e., equivalent to $\mathrm{Im}[\chi^{(3)}]$ and it is called NLMML.

In NLMML, a non-linear crystal (NLC) is inserted in the laser cavity and is placed in front of a dichroic output coupler, used in place of the usual output coupler. The dichroic mirror partially reflects fundamental wavelength (FW) but totally reflects the second harmonic (SH) beam. The FW produces SH in its first pass and if the SH beam experiences a proper phase shift with respect to the FW beam, the SH power is almost totally reconverted into FW during the second pass through the NLC. In this condition, the reflectivity of the FW increases with input power and the system shows losses decreasing with power, i.e., an equivalent negative $\mathrm{Im}[\chi^{(3)}]$. The advantages of NLMML are: (i) it is based on a reversible loss-less process, (ii) the response is instantaneous and so the pulse shortening is equally effective on both rising and trailing edges of the pulse. Picosecond regime is the ultimate short pulse (highest peak power) useful in non-linear optics without invoking non-stationary effects, if the medium lengths are properly chosen. Earlier we have reported stability study [15] and prediction of pulse width [16] of a NLMML laser, which are found to be consistent with the experimental observations. Here we report on the use of this laser for comprehensive measurement of second-order non-linearity of powdered crystalline sample.

In §2 we describe the development of a diode laser array pumped NLMML $Nd:YVO_4$ laser using type-I (xy) SHG cut (1064 nm) LBO crystal as NLC. The laser can operate in cw ML as well as in simultaneously mode-locked and Q-switched regime. The QML regime can be modified for better amplitude stability of the QML

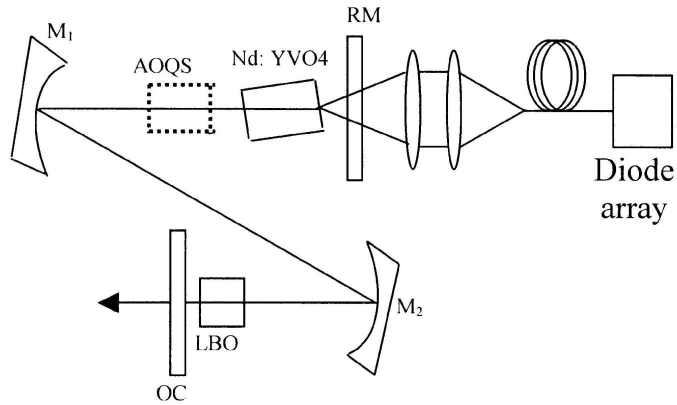

Figure 1. Schematic diagram of (actively Q-switched and) non-linear mirror mode-locked laser. OC – output coupler with 100% reflectivity at 532 nm and 78% reflectivity at 1064 nm; RM – rear mirror: the pump surface of it is coated for anti-reflection at 808 nm and the cavity surface is coated for 100% reflectivity at 1064 nm; M_1 and M_2 – curved mirrors with radius of curvature 500 and 250 mm respectively and reflectivity of 100% at 1064 nm. AOQS – acousto-optic Q-switch.

envelope by incorporating an acousto-optic Q-switch (AOQS), which provides an actively Q-switched pulse envelope and the mode-locked ps pulses due to NLM lie underneath it. In §3 we describe the powder SHG measurement for different regimes of operation of the laser and compare the results obtained.

2. Development of laser

2.1 *NLMML laser*

The schematic of the oscillator cavity is shown in figure 1. The active material is a $4 \times 4 \times 8\,mm^3$, a-cut, Nd : YVO_4 crystal having 0.5% Nd^{3+} concentration and anti-reflection coated on both faces for wavelengths 1064 nm and 808 nm. To reduce the effect of undesired reflection, the crystal is tilted by an angle of 2°, and is placed in an air-cooled heat exchanger. The crystal is end-pumped by a fiber-coupled laser diode array (Bright Solutions, Italy), emitting radiation of wavelength 808 nm and maximum output power of 15 W, through the rear mirror (RM), which is anti-reflection coated at 808 nm at the rear side and has high reflectivity (>99.5%) for 1064 nm on the other side. The pump beam coming out from the fiber of core diameter 0.6 mm and numerical aperture 0.22 is imaged on the Nd : YVO_4 crystal using two lenses of focal length 15 and 12 mm to a spot size 0.48 mm. Two concave mirrors M_1 and M_2 of radius of curvature 500 and 250 mm respectively are used to focus the beam onto the NLC, placed before the output coupler, having 100% reflectivity at 808 nm and 78% reflectivity at 1064 nm.

A 15 mm long type-I ($\theta = 90.0°$ and $\varphi = 11.6°$) cut LiB_3O_5 (LBO) (Eksma, Lithuania) crystal, anti-reflection coated at both faces for wavelengths 1064 and 532 nm, is placed very close to the dichroic output coupler. The different arms of the Z-shaped passive cavity of length 81.7 cm are optimized to be 250, 405 and 162 mm respectively for stable mode-locked operation. The TEM_{00} mode size (radius) at $Nd:YVO_4$ crystal and LBO crystal are calculated to be 0.28 and 0.084 mm respectively. A very small part of the cavity radiation (at 1064 nm) transmitted through the mirror M_1 is used to monitor the pulse shape by a silicon photodiode (UDT-HS-040, USA) of rise/fall time 0.5 ns and a 500 MHz (Tektronix TDS 3054B, USA) oscilloscope. The distance between the LBO and output coupler is optimized to control the air dispersion of the FW and SH beam by introducing proper phase shift between them and to achieve the maximum loss modulation.

The laser output power at 1064 nm was measured under both cw and mode-locking condition using a power meter (Ophir Optoelectronics, Israel, NOVA display and 30-A-SH sensor). The threshold pump power of cw lasing is 5.0 W, which is a little bit high when compared to the calculated one. This may be attributed to the inefficient coupling in the fiber end. The stable ML is obtained for a limited pump power range of 9–10 W. Due to the thermal lensing in the crystal, the mode-locking is very sensitive to variation of mode structure due to pump power variation. The laser shows a tendency to pass to QML regime. The stability of the single pulse cw ML crucially depend on the wave vector mismatch between the FW and the SH beam in the LBO crystal [15]. The repetition rate and width for Q-switched pulse are measured to be 93 kHz and 440 ns. The Q-switched pulse width is found to vary as the distance between the output coupler and the LBO crystal is changed. The pulse width is decreased to 90 ns when the distance is increased from 3 to 8 mm. Over 300 kW of peak power is thus obtained in QML regime. The repetition rate for mode-locked pulses is 178 MHz (oscilloscope measurement). The width of mode-locked pulses is measured by SHG intensity autocorrelation. A 3 mm thick type-I SHG cut ($\theta = 22.8°$) anti-reflection coated BBO crystal is used for the autocorrelation measurement. A non-collinear SHG geometry is used with non-collinear angle of 5.0°. The collimated beam spot size is sufficient enough to avoid spatial overlapping effect due to non-collinearity and walk-off effect in BBO crystal. The measured FWHM width is 8.4 ps; whereas the bandwidth limited pulse width is 1.7 ps considering the sech^2 pulse shape. Such a difference can be attributed in part to the group velocity mismatch of the 1064 and 532 nm pulses in 15 mm long LBO crystal. The group velocity mismatch parameter $\delta = 43.94$ fs/mm causes a 1.3 ps temporal delay between the FW and SH beam in a 15 mm long LBO crystal. Although this delay is smaller than the bandwidth limited value, it will have significant effect on the ultimate pulse width [5]. This is due to the fact that the SH pulse is not completely converted back during the second pass and the leading edge of the FW pulse suffers a loss which is more pronounced for shorter pulses thereby limiting the ultimate pulse duration and pulse shape deformations. The beam quality parameter (M^2) is also measured by knife-edge method. It is found that $M^2 = 2$ for output power of 1 W. But this value decreases to 1.27 as the output power increases to 3.0 W. The peak power attained in the cw ML regime is ~1.5 kW.

2.2 *Actively Q-switched NLMML laser*

The simultaneous passive Q-switching and mode-locking always suffers from poor amplitude stability especially with Nd : YVO_4 as gain medium, where the gain is very high but the fluorescence lifetime is much less. However, the advantage of getting high peak power in QML regime as well as the amplitude stability can be achieved by incorporating in the cavity a readily available AOQS operating at frequencies of several kHz. This device can lead to a stable QML regime in which the repetition rate is fixed by an external modulating signal source. We used an AOQS (Neos Technologies, USA) with faces cut for Brewster angle at 1064 nm and is driven by a radio frequency signal of 27.2 MHz with a modulation available in the frequency range 0–50 kHz. First the laser is optimized for stable Q-switched operation and then the LBO crystal is inserted to get the passive ML. The stable QML operation is found to occur for repetition rates higher than 35 kHz. The pulse width of the picosecond pulses measured by a non-collinear SHG autocorrelation employing a 3-mm long BBO crystal is found to be 9 ps. The peak power thus obtained in this regime is over 341 kW and furthermore the amplitude stability of the QML pulses is increased significantly. The measured beam quality parameter (M^2) of the output beam is 1.29 in the horizontal direction, and 1.38 in the vertical direction.

3. Powder SHG measurement

For powder SHG, we use 90° geometry to increase the sensitivity of measurement. Under this configuration, the photo-multiplier tube (PMT) is placed at an angle of 90° to the pump beam, which is incident on the sample at an angle of 45°. As the pump laser cavity contains a second harmonic generator crystal for non-linear mirror mode-locking/Q-switching, the output beam contains a slight amount of SH radiation. A visible cut but IR pass filter is used to filter out SH radiation from 1064 nm beam. The extinction of this filter at 532 nm is better than 10^{-5}. We have also verified the absence of 532 nm radiation in the main beam by allowing the light to fall on microscope glass plate without any non-centrosymmetric sample on it. No SHG signal is detected by photo-multiplier tube (PMT) in this condition. The fine powdered sample is placed in a specially made small ring-hole on a microscope-slide. As we know that the response depends on the size of the micro-crystals, we crushed all the samples to almost the same size (∼50–100 μm). Second harmonic radiation generated by the randomly oriented grain crystals is detected by a PMT (Model IP21, EMI Electronics Ltd). The sample and PMT are kept in an enclosure so that the external lights, especially the bright green (532 nm) light generated by the LBO crystal cannot enter into the PMT. We performed the powder SHG of urea and KDP in two regimes of laser operation. Figure 2 shows the oscilloscope recording of fundamental as well as the second harmonic signal for urea and KDP samples for the laser operating in simultaneous passively Q-switched and passively mode-locked regime while figure 3 shows the same for the laser operating in actively Q-switched and passively mode-locked regime. Because of the long transit time (20 ns) of the signal within PMT dynodes, it is not possible to trace the second harmonic

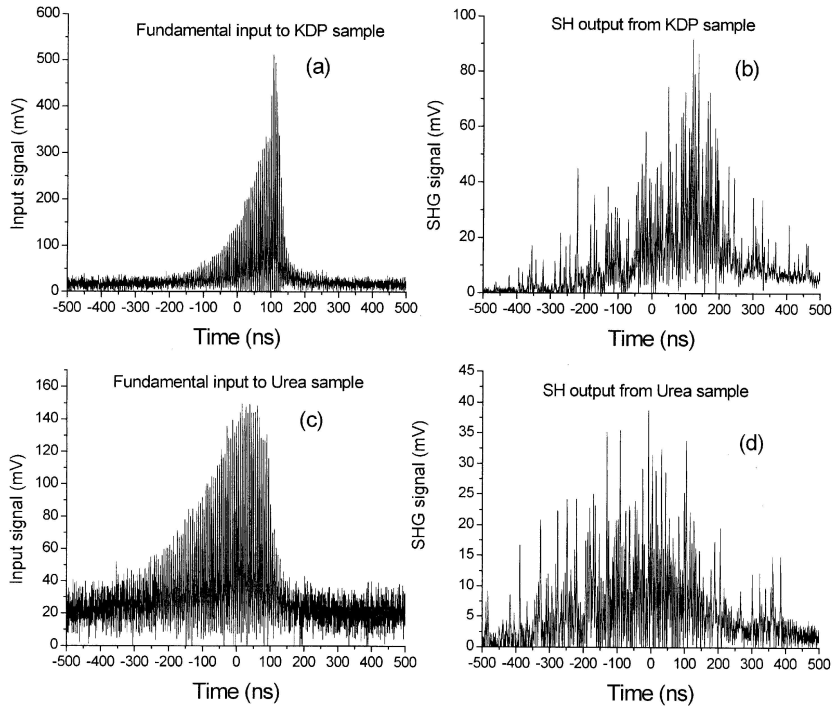

Figure 2. Oscilloscope recording of fundamental and the second harmonic signal for KDP and urea in the simultaneous passively Q-switched and passively mode-locked regime of operation of the laser.

pulse train as a faithful replica of the fundamental train. But the correspondence between the central peak of the fundamental and that of the SH is adequately distinct to measure the relative conversion. The beam radius at the sample is calculated to be 0.52 mm and the corresponding average peak intensity at 1064 nm in passively Q-switched and passively mode-locked mode incident on the sample is over 70 MW/cm^2. For the same input beam the peak intensity is over 80 MW/cm^2 for the laser operating in actively Q-switched and passively mode-locked regime. The ratio of non-linear coefficient for urea with respect to KDP is calculated from the following equation:

$$\frac{I_{\mathrm{SH}}^{\mathrm{urea}}}{I_{\mathrm{SH}}^{\mathrm{KDP}}} = \left(\frac{\mathrm{d}^{\mathrm{urea}}}{\mathrm{d}^{\mathrm{KDP}}} \times \frac{I_{\mathrm{FW}}^{\mathrm{urea}}}{I_{\mathrm{FW}}^{\mathrm{KDP}}}\right)^2 .$$

The ratio of non-linear coefficient for urea with respect to KDP ($\mathrm{d}^{\mathrm{urea}}/\mathrm{d}^{\mathrm{KDP}}$) is calculated to be 2.25 for passively Q-switched and passively mode-locked mode of operation, while the ratio is 2.9 for the laser operating in actively Q-switched and passively mode-locked regime which are very close to the measured value of 3.0 under phase-matched condition [17].

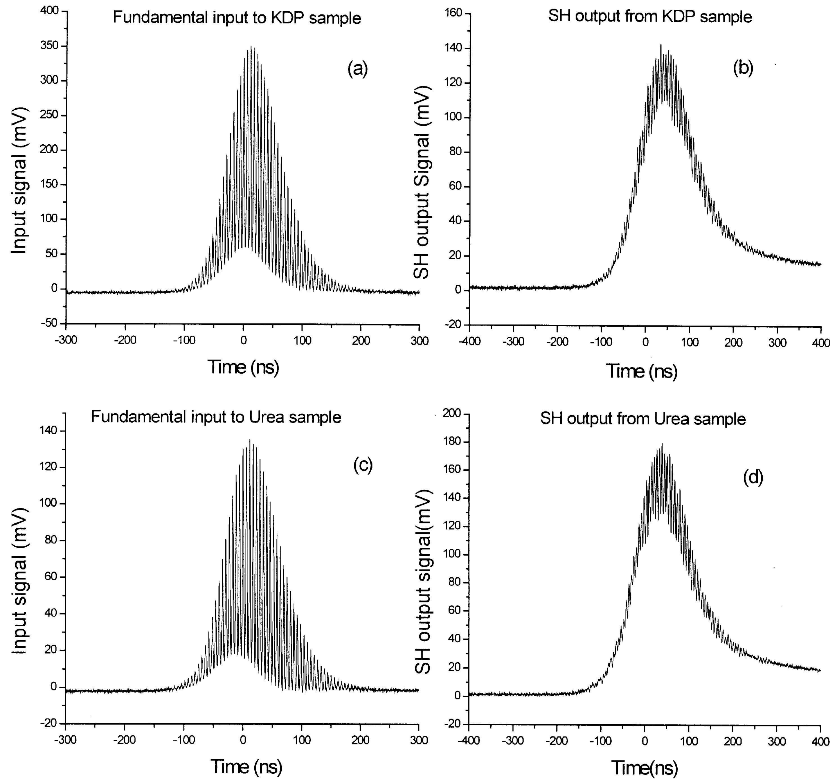

Figure 3. Oscilloscope recording of fundamental and the second harmonic signal for KDP and urea in the actively Q-switched and passively mode-locked regime of operation of the laser.

4. Conclusion

A passively mode-locked cw diode pumped Nd : YVO_4 laser has been built using the non-linear mirror as saturable absorber. The laser is operated in actively Q-switched and passively ML regime by incorporating an acousto-optic modulator for better stability of QML pulses as well as high peak power of over 340 kW. Measured value of effective non-linearity of urea with respect to KDP proves the suitability of the laser for powder SHG measurement.

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