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Non-critical phase-matched second harmonic generation in $Gd_xY_{1-x}COB$

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ABSTRACT We have characterized non-critical phase-matching (NCPM) for both Type I and Type II second harmonic generation (SHG) in y-cut $Gd_xY_{1-x}COB$ using a nanosecond optical parametric oscillator (OPO). The variation of the NCPM wavelength with temperature was investigated for different values of the compositional parameter x. Efficient SHG of 1064 nm was achieved by choosing the suitable compositional parameter $x = 0.28$ and by tuning the temperature of the crystal to 52 °C. Using a 25-mm-long $Gd_{0.28}Y_{0.72}COB$ crystal, conversion efficiencies of 41 and 43% were obtained respectively from a mode-locked Nd : YAG and a Q-switched Nd : YAG laser.

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1 Introduction

In recent years, there has been a growing demand for frequency agile or specific visible and ultraviolet laser sources in medicine, industrial processing, remote sensing, laser printing, optical displays, and other areas. At this time, the availability of laser frequencies in the visible and UV is limited by laser materials and pump sources. Thus, the reliance on nonlinear methods of frequency generation demonstrates the need for new nonlinear harmonic crystals with the ability to frequency convert a wide variety of laser wavelengths.

Rare earth calcium oxyborate $ReCa_4O(BO_3)_3$ (ReCOB, Re = Y, Gd) has attracted great attention as a new nonlinear optical (NLO) crystal for frequency generation since its earliest development. ReCOB combines some of the more attractive mechanical and optical properties in one crystal in comparison with the most commonly used NLO crystals (KDP, KTP, BBO, and LBO) [1]. ReCOB exhibits high nonlinear coefficients, a broad transmission band, a high damage threshold, and non-hygroscopic properties. In addition, the crystal melts nearly congruently, so that large single crystals can be produced by the Czochralski melt-pulling technique [2]. Furthermore, ReCOB can easily be doped with rare earths such as Yb or Nd lattice sites and, as a consequence, good lasing properties have been demonstrated [3, 4]. ReCOB has also been shown to be a promising new NLO crystal for high-average-power harmonic-conversion applications [5].

Yoshimura et al. have shown that a substitutional solid solution of $Gd_xY_{1-x}COB$ with the right composition can be used to perform non-critical phase-matching (NCPM) second harmonic generation (SHG) [6, 7]. In NCPM, phase matching is achieved along one of the principal dielectric axes of the crystal. Two major advantages result from NCPM: large angular acceptance bandwidths can be obtained and very long crystals can be used to increase the conversion efficiency because of the absence of beam walk-off. Several studies have shown how tuning of NCPM in $Gd_xY_{1-x}COB$ can lead to efficient SHG for a given wavelength: (i) by compositional tuning, demonstrated by Wang et al. [8], who measured the NCPM wavelengths for different values of the compositional parameter x; and (ii) by temperature tuning, demonstrated by Burmester et al. [9], who showed the dependence of NCPM wavelengths on temperature for Type I SHG. In this paper, we describe, for the first time to our knowledge, how combined temperature and compositional tuning of $Gd_xY_{1-x}COB$ permits efficient conversion efficiency between 723 and 831 nm for Type I SHG, and between 1022 and 1205 nm for Type II. We demonstrate this with the NCPM SHG of Nd : YAG at 1064 nm with a Q-switched laser and with a mode-locked laser.

The objective of this study was to evaluate the potential of various formulations of $Gd_xY_{1-x}COB$ crystals as nonlinear optical converters for high-power laser radiation and to confirm the ideal composition for NCPM SHG of 1064 nm, given the variation in the measured ideal compositional parameter in the literature [6, 8]. Specifically, we report the NCPM wavelengths for SHG of y-cut $Gd_xY_{1-x}COB$ crystals for different compositional parameters at room temperature. In addition, the dependence of the NCPM wavelengths with increasing temperature was investigated for the different crystals. A 25-mm-long crystal with a compositional parameter of $x = 0.28$ was temperature-tuned to frequency double both a mode-locked and a Q-switch Nd : YAG laser, achieving a conversion efficiency of over 40% in both cases.

2 Composition tuning

The crystals used in these experiments were grown by the conventional rf-heating Czochralski pulling method.

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The complete details of the crystal growth of ReCOB have been described previously [2]. Crystals with different compositions were grown by adjusting the compositional parameter x of the starting materials (Y_2O_3, CaO, and B_2O_3). ($x = 0.1$, 0.16, 0.28, 0.3, 0.31, and 0.5). The composition was adjusted during the growth to compensate both for evaporation and non-congruency. Crystals 40 mm in diameter and 200 mm in length with good optical quality were obtained. These crystals were cut in the y-direction to 25 mm in length and the ends were polished to optical flatness.

If n_x and n_z are the wavelength-dependent refractive indices in the two directions perpendicular to the direction of propagation y, the theoretical value of the NCPM wavelength λ_{NCPM} for a given composition x can be derived from the phase-matching condition given by:

For Type I:

$$n_z\,(x, \lambda_{\mathrm{NCPM}}) - n_x\left(x, \frac{\lambda_{\mathrm{NCPM}}}{2}\right) = 0\,; \tag{1}$$

For Type II:

$$n_x\,(x, \lambda_{\mathrm{NCPM}}) + n_z\,(x, \lambda_{\mathrm{NCPM}}) - 2n_x\left(x, \frac{\lambda_{\mathrm{NCPM}}}{2}\right) = 0\,. \tag{2}$$

To a first approximation, the wavelength-dependent refractive index $n_{x,z}(x, \lambda)$ for a given composition x can be obtained by a linear interpolation between the values given by the Sellmeier equations for pure GdCOB [1] and YCOB [10]:

$$n_{x,z}\,(x, \lambda) = \big(n_{x,z}\,(1, \lambda) - n_{x,z}\,(0, \lambda)\big)\,x + n_{x,z}(0, \lambda)\,. \tag{3}$$

The NCPM wavelength values were measured with a nanosecond OPO tunable from 430 to 2000 nm. The crystal was probed by 5-ns pulses at 10 Hz with approximately 10 mJ of energy per pulse. The pump wavelength was scanned over ~ 3 nm with 0.08 nm increments to find the wavelength yielding the maximum harmonic conversion efficiency. Figures 1 and 2 show the dependence of the NCPM wavelength on the compositional parameter, respectively for Type I and Type II phase matching at room temperature. The measured NCPM wavelengths were found to be between the values calculated for pure YCOB and pure GdCOB and were very close to the theoretical values calculated from the Sellmeier equations that had been reported previously [1, 10].

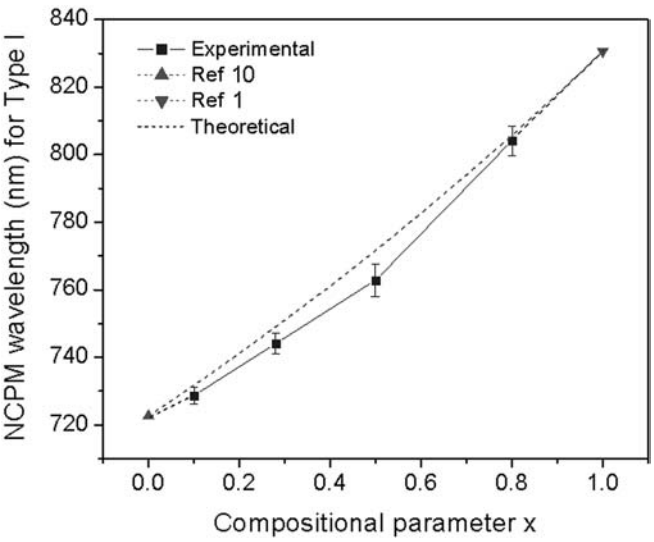

FIGURE 1 Type I NCPM wavelength for SHG in y-cut Gd_xY_{1-x}COB versus x at room temperature

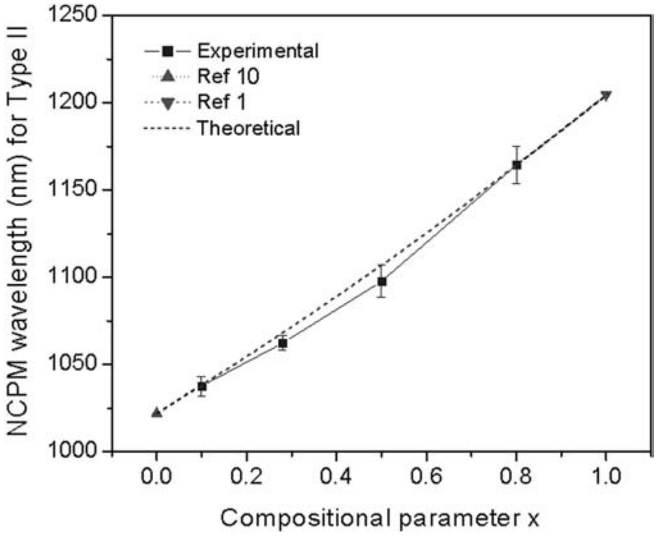

FIGURE 2 Type II NCPM wavelength for SHG in y-cut Gd_xY_{1-x}COB versus x at room temperature

These results demonstrate that SHG can be achieved at room temperature for virtually any wavelength between the NCPM wavelengths of the pure materials: 723–831 nm for Type I and 1022–1205 nm for Type II. No significant change in conversion efficiency at the NCPM wavelength was observed for the different compositions. The compositional parameter suitable for frequency doubling of 1064 nm radiation was determined to be around $x = 0.28$. This value is in good agreement with the value of 0.275 reported by Yoshimura et al. [6].

3 Temperature tuning

Temperature tuning of Gd_xY_{1-x}COB can be performed to obtain the maximum conversion efficiency at a desired wavelength close to the NCPM wavelength at room temperature. In this experiment, the shift in the NCPM wavelength with temperature was quantified. The crystal was mounted in an oven in which the temperature was gradually increased from 30 to 140 °C.

After reaching temperature equilibrium, the wavelength yielding the maximum conversion efficiency for Type I and Type II phase-matching was measured for 30, 40, 100, and 140 °C. Figure 3 shows the dependence of the Type II conversion efficiency on wavelength for different temperatures for the case $x = 0.1$. The NCPM wavelength increased with temperature without any significant change of the conversion efficiency.

In Figs. 4 and 5, the dependence of the NCPM wavelength on temperature, respectively for Type I and Type II phase matching, was studied for three different compositions of $x = 0.1$, 0.28, and 0.5. It was found that the NCPM

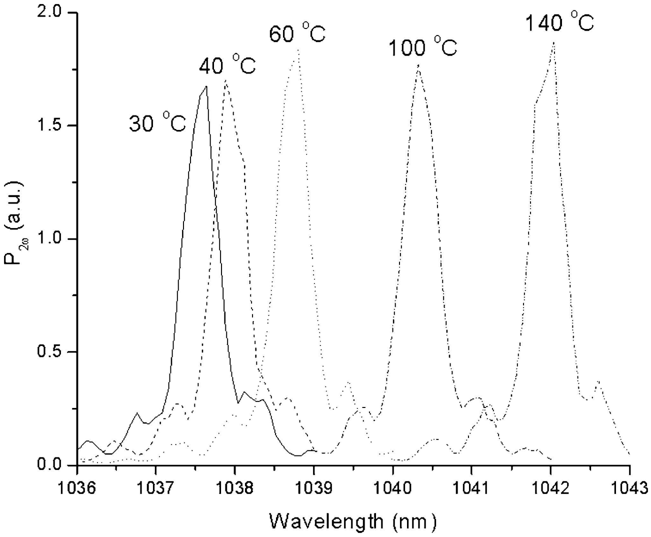

FIGURE 3 Type II second harmonic energy output vs. wavelength at different temperatures for $x = 0.1$

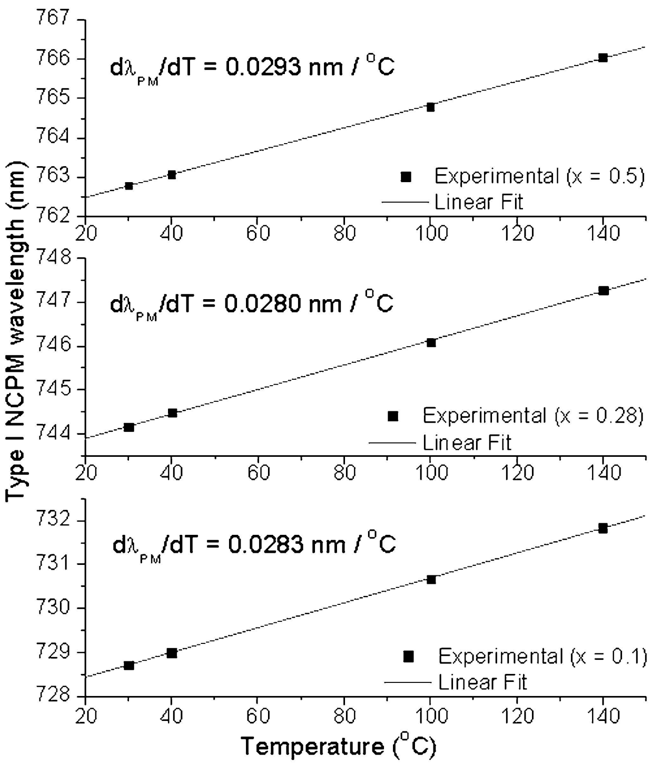

FIGURE 4 Type I NCPM wavelength for SHG in y-cut $Gd_xY_{1-x}COB$ vs. T ($x = 0.1$, 0.28, and 0.5)

wavelength followed a linear trend from 30 to 140 °C. The temperature-tuning coefficient was determined to be 0.0285 ± 0.0008 nm/°C for Type I and 0.0413 ± 0.0014 nm/°C for Type II phase matching, independent of the compositional parameter. This property could be exploited to fine-tune the crystal to match a custom wavelength close to the NCPM wavelength at room temperature, or to offset a slight deviation from the expected wavelength due to a composition error. We calculate from the temperature-tuning coefficients and from the dependence of λ_{NCPM} on x that a ΔT of 100 °C could compensate for a composition deviation of $\Delta x \sim 0.025$.

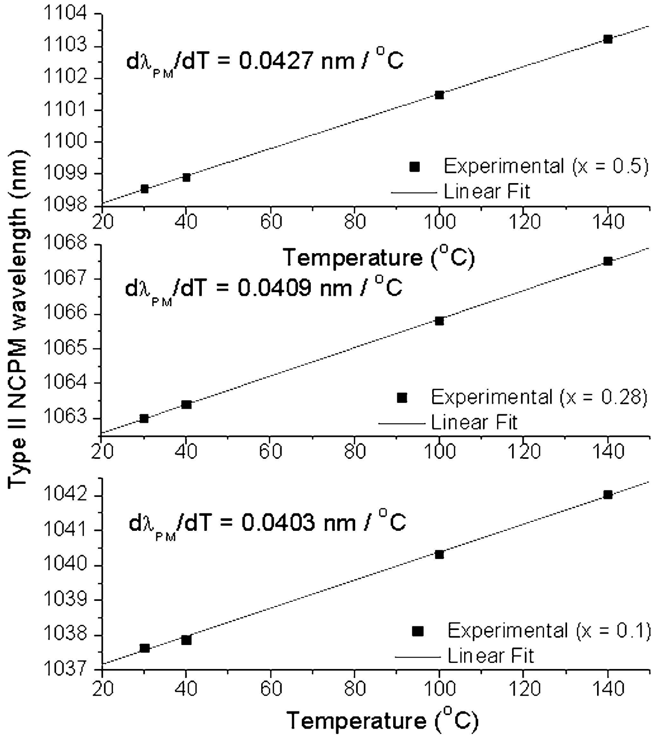

FIGURE 5 Type II NCPM wavelength for SHG in y-cut $Gd_xY_{1-x}COB$ vs. T ($x = 0.1$, 0.28, and 0.5)

4 Conversion efficiency measurement

The SHG NCPM conversion efficiency was measured for a 25-mm-long crystal with a compositional parameter of $x = 0.28$. Two SHG experiments were performed: in the first, a mode-locked Nd : YAG laser was used, delivering 35-ps pulses at 10 Hz with approximately 120 μJ of energy per pulse; in the second, a Q-switched Nd : YAG laser was used, delivering 5-ns pulses at 5 Hz with approximately 50 mJ of energy per pulse. In both cases, the optimum crystal temperature was measured to be 52 °C, which is also consistent with the results obtained from the temperature-tuning curves with the OPO (Fig. 5). Collimating telescopes were used to reduce the beam spot size such that the input peak intensity was the same and equal to 450 MW/cm^2 in both experiments. In the mode-locked experiments, 124 μJ of fundamental radiation, focused to a 1-mm-diameter beam spot (FWHM), generated 50.6 μJ of second harmonic, yielding a conversion efficiency of 40.8%. In the Q-switch experiments, 50 mJ of fundamental radiation, focused to a 1.7-mm-diameter beam spot (FWHM), generated 21.5 mJ of second harmonic, yielding a conversion efficiency of 43%. No thermal effects due to absorption of fundamental and/or second harmonic radiation were observed.

5 Conclusion

In this study we have demonstrated that efficient NCPM SHG can be achieved for any wavelength between 723 and 831 nm using Type I and between 1022 and 1205 nm using Type II phase-matching by tuning the composition of $Gd_xY_{1-x}COB$. In particular we show that composition tuning, combined with temperature fine-tuning of $Gd_xY_{1-x}COB$ offers a flexible solution to obtaining efficient frequency conversion at virtually any wavelength within 723–831 nm and 1022–1205 nm. The results in these two wavelength bands have important implications for many of today's tunable solid-state lasers: Ti:Sapphire, Cr:LiSAF, Cr:LiCAF, and Alexandrite for Type I, and Nd- and Yb-doped host materials for Type II NCPM. An ideal composition of $x = 0.28$ was confirmed for NCPM SHG of 1064-nm radiation. Furthermore, a 43% conversion efficiency was demonstrated with both a mode-locked and a Q-switched Nd : YAG laser by tuning the temperature of the crystal to 52 °C. To our knowledge, this is the highest SHG conversion efficiency reported for this crystal. A further indication from this study stems from the inherent thermo-mechanical properties of $Gd_xY_{1-x}COB$: Although high-power lasers tests were not performed in this investigation, we believe this crystal should be promising for up-converting high-average-power lasers.

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