Efficient Nonlinear Frequency Conversion to 193-nm Using Cooled BBO

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Abstract: We have developed a 193-nm laser source operating at 5-kHz that generates a near-diffraction-limited TEM_{00} beam with 35 mW average power. The conversion efficiency and stability are both significantly enhanced by cooling the BBO crystal used in the final sum-frequency mixing stage.

OCIS codes: (140.0140) Lasers; (140.7240) Ultraviolet lasers; (190.0190) Non-linear optics; (190.4140) Parametric processes.

1. Introduction

Solid-state sources of sub-200 nm light are becoming increasingly useful for a wide variety of disciplines, including material processing, photochemistry, and optical metrology. The present source in particular was designed to match the actinic wavelength of the ArF excimer laser for semiconductor photolithography metrology and small-field exposure applications. A major challenge to attaining reliable, high-average-power sources in this region of the spectrum is presented by the dearth of nonlinear crystalline media with sufficient transparency and birefringence. Several sub-200-nm laser sources have been demonstrated to date based on nonlinear frequency conversion of high-power Q-switched infra-red lasers. An early system generated 193-nm light by using cesium-lithium-borate (CLBO) to frequency-mix the fifth harmonic of a Nd:YAG laser with the 2-micron signal wave of a 1064-nm pumped OPO [1]. More recently, a 1-W sub-200-nm source at 5-kHz was demonstrated, again using CLBO to mix the output of a 45-W Nd:YLF MOPA with a 3-W (tunable) $Ti^{3+}:Al_2O_3$ laser [2]. The crystal beta-barium-borate (BBO) is of interest because its increased birefringence permits phase matching of a wider variety of optical processes, and in particular the mixing of the Nd:YAG fourth harmonic with 700-nm light to produce 193-nm light. However, the intrinsic absorption edge of BBO near 6.5 eV causes significant absorption of sub-200-nm light. This absorption, and the resulting thermal variations and crystal degradation have severely limited the usefulness of BBO for short-wavelength generation [3].

Early experiments (*ca*. 1988) on the absorption of light near the absorption edge by cooled BBO crystals indicated that the absorption increases upon cooling [4]. In stark contrast, however, more recent experiments have shown that the absorption of BBO can be greatly *reduced* by crystal cooling [5]. In particular, with high-purity Cz-grown BBO, Kouta and Kuwano showed that absorption at 193-nm exponentially diminishes from 1.4 cm^{-1} at 295K, to 0.29 cm^{-1} at 91K. While these authors presented the variation of the phase matching angle for the process $3\omega + \omega \rightarrow 4\omega$, no comparison of the high- and low-temperature conversion efficiencies was reported.

In this paper, we present a direct comparison of the performance of cooled and room-temperature BBO for nonlinear frequency mixing. We show that the generated 193-nm power level almost doubles, *ceteris paribus*, as a result of cooling the BBO to approximately 226K. Furthermore, strong thermal hysteresis effects observed at room temperature are greatly ameliorated upon crystal cooling and temperature stabilization.

2. Laser Architecture

Light at 193.4 nm is generated by frequency-mixing a 266-nm ultraviolet (UV) beam with a 708-nm infra-red (IR) beam. Each of the interacting beams is generated from a common green pump laser, a Lightwave Electronics Q-201 diode-pumped, intracavity-doubled Nd:YAG laser operating at 5 kHz, as shown in Fig. 1. This laser generates 15-ns-wide (FWHM) pulses with energy up to 2.5-mJ/pulse at 532 nm, giving an average power of ~ 12.5 W. The UV beam at 266 nm is the second-harmonic of the pump laser light and is generated by Type-1 phase matching in a 5x5x10 mm CLBO crystal. The IR beam at 708.6-nm is the signal wave of a 532-nm-pumped OPO. This OPO employs a volume Bragg mirror (VBG) output coupler to reduce the frequency bandwidth to ~1 cm^{-1} (30 GHz). The IR and UV beams, each with power levels ~1 W, are combined using beam combiner BC. The flux-grown BBO crystal is uncoated and contained in a dry atmosphere within a sealed housing. A four-stage TEC (Melcor, NJ) is used for crystal cooling; we were able to cool to a minimum crystal temperature of 223K. The 193-nm light is separated from the other wavelengths by use of a Pellin-Broca prism, and propagates through ~1m air prior to the power monitor. The 193-nm beam generated using both high- and low-temperature BBO crystals is nearly

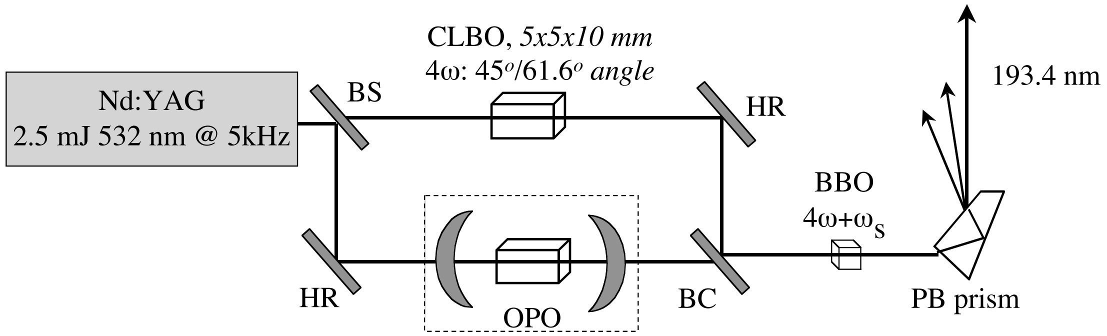

Fig. 1 Schematic Diagram of the 5-kHz 193-nm laser system. Components are described in text.

diffraction-limited (M^2 values < 1.2 in both directions) with a TEM_{00} spatial mode, and has a frequency bandwidth, as deduced by fringe analysis using a Michelson, of 2 cm^{-1} (56 GHz) with a Gaussian spectral distribution.

3. Variation of phase matching angle with temperature

We first measured the phase matching angle as a function of crystal temperature, for fixed UV and IR wavelengths, for a BBO crystal cut at 78.5 degrees. Low powers were applied to reduce absorption-induced thermal effects. The change in external angle (the required crystal rotation angle) relative to room temperature is plotted in Fig. 2 for temperatures down to ~253K. The slope of the resulting curve is 0.93 mrad/K. Using an average index of refraction of 1.7 ($n_o(266) = 1.76$, $n_o(710) = 1.66$, $n_e(193) = 1.73$) this corresponds to a variation in the internal phase-matching angle of ~0.55 mrad/K. This value is about an order of magnitude greater than that predicted by the SNLO computer code based on available thermal coefficients [6]. Extrapolation of the curve from these data to 225K suggested a crystal cut angle of 76 degrees; such a crystal was then ordered (Ekspla, Vilnius, Lithuania) and used for the subsequent data. This cut angle proved to be nearly exactly correct (within +/- 0.25 degrees).

4. High-power operation at room temperature

Power generation and phase-matching properties were first investigated using a room-temperature, 78.5-deg-cut 8x8x6-mm BBO crystal. From 800 mW of 266-nm light and 1.2-W of 710-nm light incident on the crystal (each beam had a TEM_{00} mode, ~300 μm FWHM diameter), over 20 mW of 193-nm light was measured. The high absorption of the generated 193-nm light (the 6-mm crystal transmission is ~40%) was manifested in dramatic hysteretic phase-matching behavior, as shown in Fig. 3. Closed data points were taken for increasing crystal angle, and open data points for the opposite direction. Each data point represents the final power value that stabilized after 20 sec; each error bar indicates the initial power value achieved immediately upon crystal rotation. The absorption causes several problems: first, depending on the direction of crystal rotation, the temperature variation of the crystal either acts to bring the nonlinear process closer to the phase-matched condition, or to further detune it. Secondly, the optimum angle changes dramatically over time. The effective angular tolerance is difficult to determine precisely, but is ~1 mrad external, corresponding to 0.353 mrad-cm internal. This value is 1.8x that predicted by SNLO. The causes for this discrepancy are under investigation, but may be related to a thermal-induced dephasing.

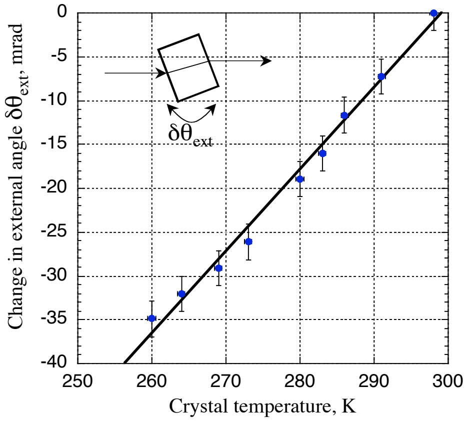

Fig. 2. Variation in external phase-matching angle with BBO crystal temperature.

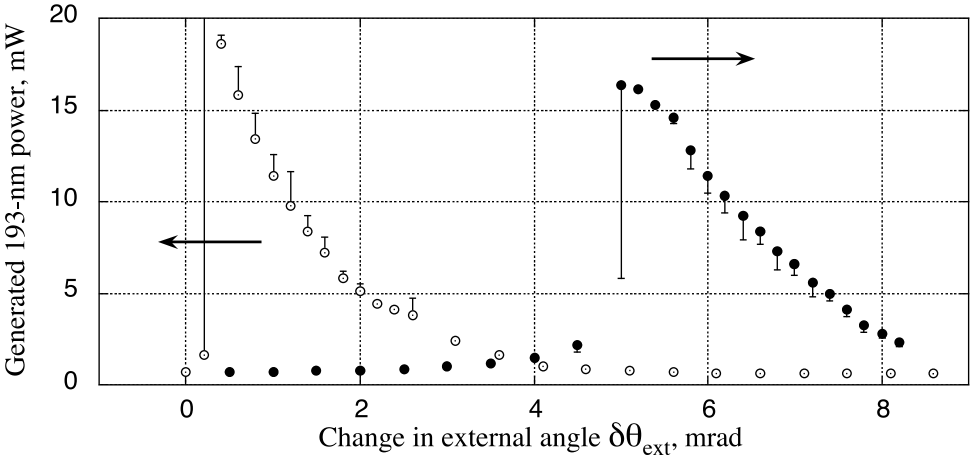

Fig. 3. Hysteretic phase-matching curve, room-temperature 78.5-degree-cut 8x8x6-mm BBO crystal.

5. High-power operation at 226K

A 76-degree-cut 8x8x6 mm BBO crystal was then cooled to 226K and the experiment repeated with all other factors constant. The transmission of the cooled crystal was not measured. Significantly, many beam parameters (spatial mode, rms stability, and pulsewidth) remained the same, but the maximum output power increased to just under 40 mW (~4% efficiency). The observed phase matching curve is shown in Fig. 4. Mechanical backlash was eliminated prior to reversing the crystal rotation, so that the observed hysteresis is due entirely to crystalline heating effects. The hysteresis is much reduced relative to the room-temperature case, and the temperature-stabilized crystal exhibits much greater long-term power stability. Interestingly, the angular tolerance remains the same (~1 mrad external) as the room-temperature case. The fact that the average heat dissipation in the crystal (the product of the 193-nm power and absorption) is also nearly the same suggests that thermal effects may be the cause of this behavior.

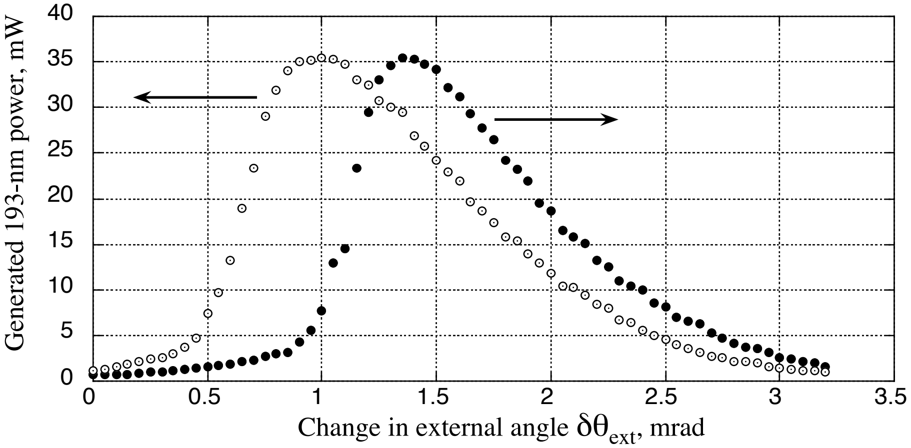

Fig. 4. Phase-matching curve of a 76-degree-cut 8x8x6-mm BBO crystal cooled to 226K.

6. Conclusions

Cooled BBO was used to nearly double the nonlinear conversion efficiency to 193-nm. The discrepancy between predicted and observed behavior is likely due to thermal effects and is the subject of further investigation.

Acknowledgements

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