Efficient second harmonic generation in β-barium borate by a diffraction-limited copper vapor laser

R. Pini, R. Salimbeni, G. Toci, and M. Vannini

The diffraction-limited beam of a copper vapor laser employing a self-filtering unstable resonator was used to induce second harmonic generation in a nonlinear crystal of β-barium borate. Despite the moderate emission characteristics of our small-scale laser device (1.5-W average power, 25-kW peak power at 511 nm), we obtained average and peak power conversion efficiencies of approximately 20 and 30%, respectively, which improved on the previously reported results by a factor of 2.

Key words: Copper vapor laser, self-filtering unstable resonator, second harmonic generation, β-barium borate.

Introduction

After approximately 25 years of technological development, copper vapor lasers (CVL's) have reached a high average power (up to 100 W), a high pulse rate (5–20 kHz), and a relatively high efficiency (1%), leading to new scientific and industrial applications for these lasers. The field of applicability of this class of lasers could be extended by frequency conversion to the ultraviolet region. Second harmonic generation (SHG) in nonlinear crystals of the 511- and 578-nm emission wavelengths could favorably compete with the presently available ultraviolet sources, if the overall efficiency of this process could become consistently high.

To our knowledge, only a few studies have been reported on this subject.[1,2] Kuroda *et al.*[3] demonstrated a conversion efficiency in β-barium borate (BBO) of ~9% of the average pumping power by using a high-quality beam, corresponding to an instantaneous conversion at the peak of 17%. According to them, this large difference was due to the complex pulse-shape structure and to the observed time evolution of the beam quality during the pulse, which is an intrinsic characteristic of the positive-branch unstable resonator they used. Observing similar beam-quality evolution, Freegarde and Naylor[4] reported SHG in BBO, leading to a substantially lower conversion at the pulse onset with respect to the tail; the average power conversion efficiency was 11%. Naylor *et al.*[5] pumped BBO with a high-power oscillator-amplifier CVL. They obtained lower efficiencies but generated some thermal effects in the crystal arising from the high-power deposition, such as thermal detuning or even catastrophic damage.

For the efficient SHG of CVL radiation, we must fulfill certain performance criteria with the laser source. First, we must conduct a high-efficiency CVL operation, which requires great attention to the choice of the operating parameters to maximize both the pulse peak power and the average power. Second, we must obtain a high-quality diffraction-limited emission without substantially reducing the output power to permit high-intensity focusing into the crystal. Third, we must find the suitable temporal shape of the laser pulse best tailored to the peak power level.

In previous work[6] we investigated a CVL operation with a self-filtering unstable resonator (SFUR). Because of the high gain of the laser medium, this resonator configuration permits diffraction-limited operation (110 μrad at 511 nm in a 13-mm beam diameter) with 50% average power extraction (2.5 W on both emission lines with respect to 5 W of the stable resonator) and 100% peak power extraction (25 kW). In addition, the SFUR intrinsically provides diffraction-limited emission from the onset of the pulse, whereas in most of the unstable resonators the beam divergence improves during the pulse evolution. The pulse shape is well characterized by two main peaks arising from the gain-saturation effect that is induced in the active medium by the intracavity-aperture feedback. The laser beam comes out collimated and its spatial intensity distribution is

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almost flat (except for a central well caused by the hole of the scraper mirror), implying saturation of the gain in the wings of the original Gaussian distribution. In principle, all of these features can enhance the efficiency of SHG.

The present investigation consisted of two phases. In the first phase, in the absence of thermal effects that can disturb the phase-matching condition, we considered the limits of the conversion efficiency of a single pulse with the pulse energy and peak power generated in our pumping scheme. For these measurements, the pump repetition rate of the pump beam was externally reduced by means of a chopper. This reduction resulted in a low-average power level coupled into the crystal. In the second phase, in search of thermal disturbances that might reduce the efficiency, we measured the SHG conversion efficiency at the maximum average power (1.5 W) of the green emission line available from our laser device.

Adaptation of the Classical SHG Theory

The top-hat transverse distribution of the SFUR beam described above does not permit us to use directly the classical SHG theory of Boyd and Kleinman,[7] which evaluates the optimum conversion parameters for a Gaussian distribution. In our case, we have assumed a focused plane-wave distribution with a limited aperture and have developed a simple modification of the original Boyd–Kleinman theory to fit the present coupling geometry better. If we approximate the field distribution of the focused plane wave of finite aperture in the close proximity of the focus with a Gaussian distribution, we can relate the typical Gaussian parameters to those of the plane wave as follows:

$$w_o = 2\sqrt{2}f/(r_o k_1), \quad (1)$$

radius of the spot focal at $1/e^2$,

$$b = 8f^2/(r_o^2 k_1) = k_1 w_o^2, \quad (2)$$

confocal parameter, where r_o is the radius of the plane-wave beam on the lens, f is the focal lens, and k_1 is the wave number. This approximation is valid if we neglect second-order terms of r/w_o and for $|z/b| \leq 2$ (z being the distance from the focus along the optical axis), which is a reasonable assumption in our case because the crystal is set at the focus and its length is comparable with the confocal parameter. Thus, the second harmonic power under optimized phase-matching conditions can be calculated by means of the Boyd–Kleinman theory, but with the introduction of our modified Gaussian parameters. In particular, the optimum focal length has the following expression:

$$f_o = [Lk_1 r_o^2/(8n_1\xi_m)]^{1/2}, \quad (3)$$

where $\xi_m = L/b = 1.392$ is the optimized strength-of-focusing parameter[7] and L is the crystal length. We note that f_o calculated for the focused plane wave is $\sqrt{2}$ times smaller than that of the Gaussian beam (obtained by replacing r_o with the radius at $1/e^2$ of the Gaussian distribution on the lens). Introducing numbers typical of our system ($L = 6.5$ mm, $r_o = 6.5$ mm, $\lambda = 511$ nm), we obtain $f_o = 42.5$ cm; we can also evaluate the conversion factor $C = P_2/P_1^2 = 3.16 \times 10^{-5}$ W^{-1}. It is important to remember that both the Boyd–Kleinman theory and the present modification are valid only in the limit of the undepleted pump.

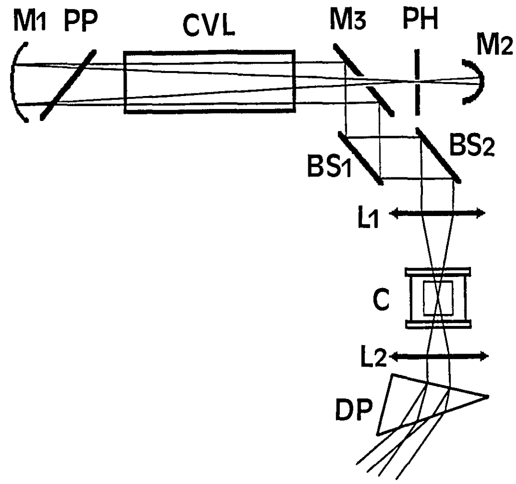

Fig. 1. Experimental setup: M_1, M_2, M_3, 3-m and 5-cm-radius mirrors and scraper mirror, respectively; PH, pin hole (200 μm in diameter); PP, plate polarizer (at 511 nm); BS_1, BS_2, beam splitters (96% reflectivity at 511 nm at 45°); L_1, L_2, coupling and collimating lenses, respectively; C, cell and BBO crystal; and DP, dispersing prism.

Experimental Setup

The setup is shown schematically in Fig. 1. The green component of the SFUR beam was almost totally polarized in the horizontal plane by the intracavity-plate polarizer set near the rear resonator mirror. Two dichroic beam splitters were used to separate the 511-nm emission line. The BBO crystal used in the experiment was 4 × 5 × 6.5 mm (from CSK) with uncoated faces and was cut at 51° with respect to the optical axis for the 511-nm wavelength conversion; it was sealed in a cell with antireflection-coated quartz windows and was located on a 5-axes micropositioner stage for fine adjustments. Lens L_1 coupled the fundamental beam into the crystal, lens L_2 collimated the output, and an ultraviolet-grade uncoated quartz prism separated the fundamental and the harmonic beams. During the first phase of investigation (conversion efficiency measurements at low-average power) we set a chopper before the crystal cell to reduce the pumping-repetition rate from 5 kHz to 30 Hz. This reduction allowed us to perform direct measurements of the energy of each single pulse with a Laser Precision RJP-735 energy meter. Temporal shapes were detected by means of a fast vacuum photodiode connected to a 0.5 GHz band-width oscilloscope. The average power was measured by a Scientech 38-0201 pyroelectric monitor and an EG&G 460-2 silicon detector.

Measurements and Results

Different focal lengths of the coupling lens, ranging from 20 to 100 cm, were tested to optimize the SHG efficiency. During the experiments, the average power of the 511-nm beam ranged between 0.9 and 1.5 W as measured before the coupling optics. After the fine-alignment procedure was completed, a bright ultraviolet beam at 255 nm was available for detection and measurements. The net energy of the fundamental pulse entering the crystal and that of the second harmonic pulse were evaluated by measuring the energy of these two spectral components after the dispersion of the prism, taking into account their losses through the optical components involved. Thus, careful preliminary measurements of the transmission of each optical component at both fundamental and second harmonic wavelengths were performed with a spectrophotometer, giving 96% transmission at 511 nm for the cell input window, 6% reflection on each uncoated crystal face, 91 and 94% transmission at 511 and 255 nm, respectively, for the cell output window, and 90% transmission at both wavelengths for the collimating lens L_2. The transmission of the prism was measured directly for the horizontally polarized pump and for the vertically polarized second harmonic beam and was found to be 97 and 87%, respectively.

In the measurements at low-average power, we evaluated the SHG conversion efficiency in terms of energy efficiency η_E, representing the ratio between the energy of the second harmonic and fundamental pulses, and peak power efficiency η_P, as the ratio between their peak powers, obtained from the analysis of their temporal shapes.

The energy of the input fundamental pulse was measured by lowering the crystal cell to permit the beam to pass through the same optical components (cell windows included) but not through the crystal. Because of the asynchronous operation of the chopper with respect to the laser-repetition rate, each energy measurement was performed by recording the peak energy value over 100 pulses, averaging five series of measurements.

Table I reports the best values recorded of both η_E and η_P for different focal lengths. Figure 2 displays the peak power efficiency η_P as a function of the strength of focusing ξ. Note that energy and peak efficiencies as high as 21 and 33% were obtained, even

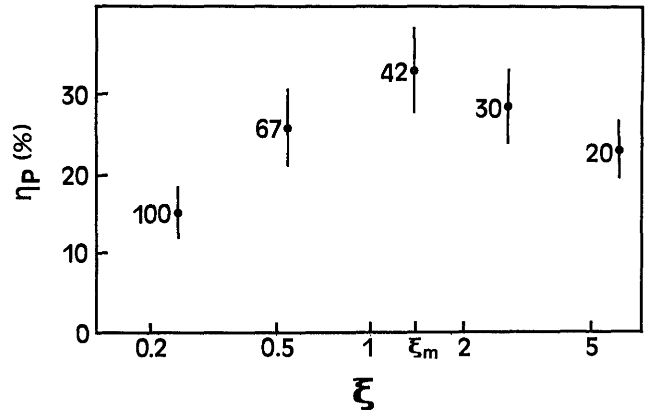

Fig. 2. Peak power conversion efficiency η_P as a function of the strength-of-focusing parameter $\xi = L/b$, corresponding to different focal lengths of the coupling lens; $\xi_m = 1.392$ corresponds to the optimum coupling conditions, according to the Boyd–Kleinman theory.

at a moderate peak power level of ~20 kW. Looking at the conversion factor $C = P_2/P_1^2$, which permits us to compare the conversion rate at different input powers, we see that the optimum focal length is 42 cm. This value, even if obtained in depleted pump conditions, appears to be in good agreement with our analytical predictions, which strictly apply to the low conversion limit.

With this focal length we performed a measurement of η_P by varying the peak power of the pump beam with a set of calibrated neutral filters. The

Table I. SHG Parameters for Different Focal Lengths[a]

Focal Length (cm)	E_1 (μJ)	E_2 (μJ)	η_E (%)	P_1 (kW)	P_2 (kW)	η_P (%)	C (10^{-5}/W)
100	178	11	10	27	4.1	10	0.55
67	138	24	17	23	6.0	26	1.12
42	118	25	21	22	7.1	32	1.51
30	129	25	19	23	6.6	28	1.12
20	170	26	15	31	7.0	23	0.72

[a] E_1 and E_2 are the energies of the fundamental and second harmonic pulses, respectively; P_1 and P_2 are their peak powers; η_E and η_P are the energy and peak power efficiencies, respectively; C is the conversion factor, defined as P_2/P_1^2.

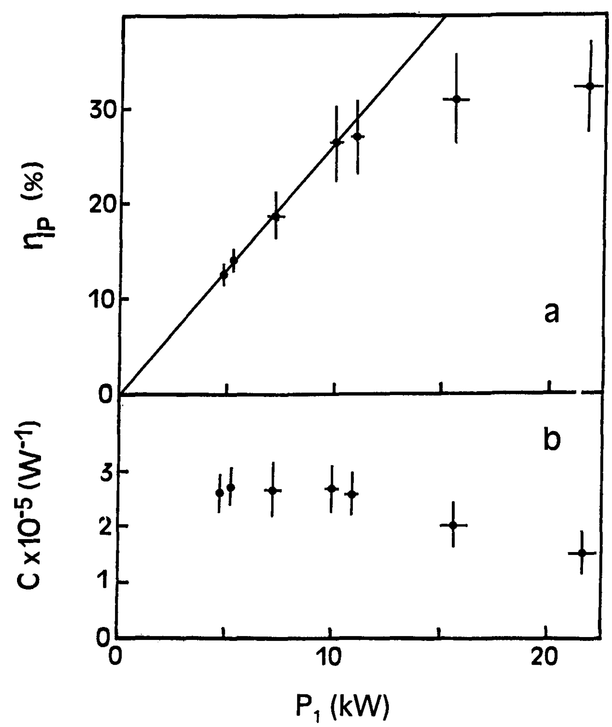

Fig. 3. Peak power conversion efficiency η_P versus pump peak power P_1 for the 42-cm focal-length lens; the straight line indicates the behavior in undepleted pump conditions. (b) Conversion coefficient $C = P_2/P_1^2$ versus peak pump power P_1.

results are shown in Fig. 3(a). Above a power level of ~10 kW the SHG process experiences a saturation effect, which is likely caused by pump depletion and some parasitic processes (i.e., two-photon absorption and self-focusing) whose occurrence was not investigated. Figure 3(b) shows the behavior of the conversion coefficient C: far from the saturation region, its value is almost constant at $2.6 \times 10^{-5}\ W^{-1}$, resulting in reasonable agreement with the predicted value of $3.16 \times 10^{-5}\ W^{-1}$.

The temporal shapes of the fundamental input, the depleted fundamental output, and the second harmonic pulse in optimized focusing conditions are shown in Fig. 4. Comparing Figs. 4(a) and 4(b) we see that the two peaks appear to be largely reduced, whereas in the second harmonic pulse of Fig. 4(c) they appear to be steepened by the nonlinearity and slightly narrowed with respect to the pump pulse. To check that the whole pump pulse undergoes an efficient conversion, we analyzed the instantaneous behavior of both η_P and C resolved for time intervals of 1 ns during the pulse evolution. The first graph, Fig. 5(a), indicates a good conversion even during the onset of the pulse, which confirms indirectly that the SFUR provides a stable high-quality beam over the whole pulse duration; the second graph, Fig. 5(b), shows that the value of C corresponding to the peaks is lower, as we expected because of the observed saturation effect.

In the second phase of measurements, by removing the chopper that externally reduced the repetition rate of the laser, we tested the SHG at the maximum average power (1.5 W) of the green emission line of our laser device. In the optimum focusing condition with the 42-cm focal length lens, we observed a maximum value of the second harmonic average power of 230 mW. Accounting for transmission losses of the coupling optics, we find that this average corresponds to an efficiency of 19.2%, which withn the experimental uncertainties compares well with the η_E value of 20.8% obtained at low-average power. The stability of the second harmonic power was tested with a photodiode, showing a fluctuation from pulse to pulse of less than 5%, generated by fluctuations of the same order of the laser emission. The same photodiode was used with a shutter to check whether the first thousand laser pulses could cause a reduction of the efficiency, induced by the thermal heating of the irradiated volume of the crystal perturbing the optimum phase matching. No evidence of such an effect was ever observed.

During the reported measurements the single-pulse intensity and the average power density at the focus inside the crystal reached maximum values of 6

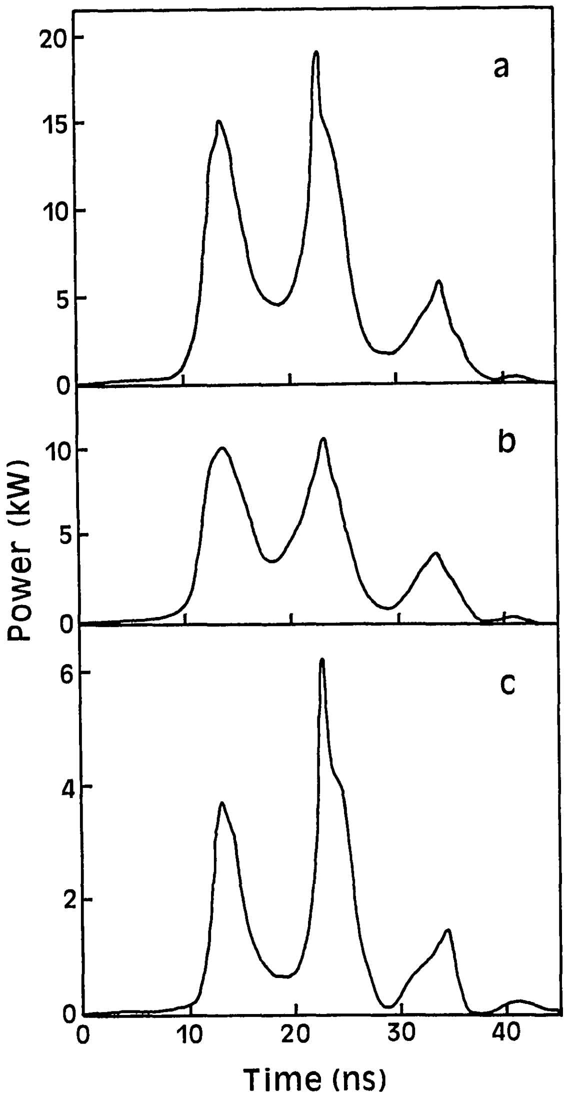

Fig. 4. Temporal shape of the (a) input pump, (b) depleted pump, and (c) second harmonic pulse.

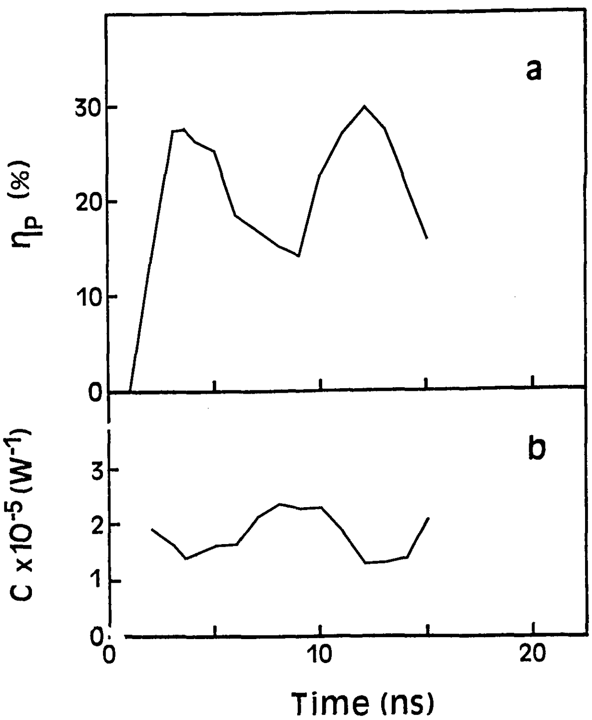

Fig. 5. (a) Instantaneous peak power efficiency and (b) instantaneous conversion coefficient during the pulse evolution; both curves are resolved for time intervals of 1 ns.

gW/cm^2 and 400 kW/cm^2, respectively; at these levels crystal damage induced by internal optical breakdown or thermal stress never occurred. In contrast, damage of the coated input window of the BBO cell was once observed during adjustment procedures with the shortest focal length lens.

Conclusions

In conclusion, we demonstrated a high SHG conversion efficiency by pumping a crystal of BBO with the CVL green emission line. Considering that these values have been achieved with a small-sized and moderate peak power laser, we believe that they represent a substantial improvement over the previously reported values. The high quality of the beam maintained throughout the whole temporal duration of the laser pulse permitted stable and high-intensity focusing into the crystal.

At low peak power levels the linear increase of the efficiency and the optimized coupling parameters predicted by the theory have been reasonably verified, whereas at higher power levels the process showed a marked saturation effect, which has to be considered to design the most favorable cost-effective coupling geometry.

The extension of these results to higher power regimes should account for possible thermal effects that reduce the intrinsic conversion efficiency, which, at the maximum average power level of 1.5 W the green emission line available from our laser device, were not observed.

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Note added in proof: A relevant recent work on SHG in BBO excited by a CVL has come to our attention after the submission of this paper. It reports similar conversion efficiencies obtained at higher average power by pumping with an injection-seeded CVL: W. Molander, M. Norton, and J. Chang, in *Conference on Lasers and Electro-Optics,* Vol. 10 of OSA 1991 Technical Digest Series (Optical Society of America, Washington, D.C., 1991), paper CTUW11.

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