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Efficient sum-frequency and second harmonic generation in a two-pass copper vapour laser amplifier

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***Abstract*. New results are presented on the efficient generation of UV radiation by using nonlinear DKDP and BBO crystals and a two-pass copper vapour laser amplifier with the enhanced peak power. The average power (average optical efficiency) of laser radiation at the sum frequency ($\lambda = 0.271$ μm) was 3.6 W (24 %) for the BBO crystal and 2.1 W (14 %) for the DKDP crystal. The maximum average second-harmonic power generated by using the BBO crystal was 3.4 W (44 %) at 0.289 μm and 2.1 W (27 %) at 0.255 μm.**

Keywords: *ultraviolet radiation, nonlinear crystal, multipass amplifier, copper vapour laser.*

1. Introduction

The application of copper vapour lasers and amplifiers for nonlinear generation of UV radiation is of great practical interest. However, they can be competitive with lasers of other types, for example, solid-state lasers [1, 2] probably at output UV powers exceeding 5 – 10 W and also at a sufficiently low cost of a watt of output radiation. This can be achieved by using comparatively cheap sealed copper vapour lasers [3, 4] emitting 10 – 25 W of output radiation. The achievement of high nonlinear conversion efficiencies and the high efficiency of the system as a whole is hindered by a number of physical reasons related to the properties of copper vapour lasers and nonlinear crystals.

A copper vapour laser (CVL) emits at the wavelengths 0.51 and 0.578 μm, which allows the second harmonic generation (SHG) at 0.255 and 0.289 μm by using nonlinear crystals DKDP, BBO, CLBO, et al. and also the sum-frequency generation (SFG) at 0.271 μm.

Unfortunately, the output radiation of usual CVLs with an unstable resonator consist of several beams formed after successive round-trip transits of spontaneous radiation in the optical resonator during a short time of inversion existence [5 – 7]. These beams have different divergences, which hinders their simultaneous use for nonlinear frequency conversion [8]. The beam formed after the last round trip of radiation in the resonator can have a low, diffraction-limited divergence; however, it contains only a small part of the laser radiation energy [2 – 10]. Because the optical efficiency of laser frequency conversion in nonlinear crystals considerably depends on the radiation power density and divergence of the beam in a crystal [11], only the output beams with nearly diffraction-limited divergence can be efficiently used for frequency conversion.

Beams with a high divergence are usually ‘filtered’ by means of a spatial filter, and the optical efficiency of nonlinear frequency conversion is determined with respect to the rest of the laser radiation incident on a crystal. In this case, the average UV output power of unstable-resonator CVLs with ADP [12] and BBO [13] crystals were 5.5 and 230 mW, respectively, while the optical efficiency did not exceed 8 % – 9 %. In papers [12, 13], a beam with a circular cross section was focused into a nonlinear crystal by spherical lenses. Later (see, for example, review 14]), considerable progress was achieved due to an increase in the fraction of low-divergence output radiation of CVLs and the use of cylindrical lenses for focusing radiation into a crystal. For example, the output power and efficiency of a 20-W CVL with a BBO crystal were 1.75 W and 34 %, respectively, at a wavelength of 0.255 μm; 1.22 W and 29 % at 0.289 μm; and 1.5 W and 19 % at 0.271 μm [15].

In [16], spherical concave mirrors mounted at an angle of 45° were used instead of cylindrical lenses, and 1.4 W of the SHG power and the 28 % efficiency were achieved at 0.255 μm. In a DKDP crystal, 0.75 W of the SFG power for the 12 % efficiency was achieved by using the optimised spherical optics producing a parallel beam of circular cross section in the crystal [10].

In the case of higher-power unstable-resonator CVLs (50 – 90 W), the SHG power at 0.255 μm noticeably increased, but the optical efficiency remained the same. Thus, the best results were 3.9 W (the 29 % efficiency) for a BBO crystal and 4.7 W (35 %) for a CLBO crystal [17]. In [18], SHG was obtained in the green spectral region by using one BBO crystal (3.2 W, 20 %) and two BBO crystals (5.13 W, 16 %). Note that in papers [12 – 18] only a fraction (0.2 – 0.5) of the total CVL power was incident on crystals, which could be efficiently converted to UV radiation.

The more efficient conversion of laser radiation can be achieved by using a scheme consisting of a master oscillator and a copper vapour laser amplifier, because in this case almost total output power can be concentrated in a diffraction-limited beam. For the 20-W amplifier, ~ 1 W of the output power and ~ 5 % efficiency were obtained at

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0.271 μm by using a KDP crystal [19], while for the 4.5-W amplifier, 0.59 W and 25 % at 0.255 μm; 0.53 W and 27 % at 0.289 μm; and 0.56 W and 14 % at 0.271 μm were obtained by using a BBO crystal [20]. In [21], a higher-power (~ 40 W) amplifier and a BBO crystal were used to obtain 3.6 W and 20 % at 0.255 μm; 2.0 W and 22 % at 0.289 μm; and 3.8 W and 14 % at 0.271 μm. The maximum SHG powers at 0.255 μm in a BBO crystal, 9 W (8 % efficiency) [22] and in a CLBO crystal, 15 W (28 %) [23], were achieved using amplifiers with the output power 113 and 102 W, respectively. In these papers, approximately half the output power of amplifiers was used for SHG and almost total their power for SFG.

Experimental studies have shown that the power of converted UV radiation increased with increasing the output power of CVLs, but the average optical efficiency remained equal to ~ 30 %. The authors of papers [14–19] assume that this restriction of the optical efficiency for most of the crystals (except CLBO) is explained by absorption of UV radiation in them, which produces the inhomogeneous heating of the crystal (the so-called thermal self-action appears [11]). It is obvious that except thermal self-action, the optical efficiency could be limited by a high divergence (even diffraction-limited) in the principal plane of the crystal if it were greater or comparable to the angular phase-matching width of a nonlinear crystal.

To increase the UV radiation generation efficiency with the use of low-power lasers, a scheme consisting of a master oscillator, a multipass amplifier, and a nonlinear crystal was proposed [24]. A multipass amplifier of a special design [25] can provide a considerable increase in the peak power of output pulses by preserving the average power at the same level. This allows one to increase the conversion efficiency in a nonlinear crystal due to high peak powers of radiation incident on a nonlinear crystal without drastic narrowing the beam diameter (or its linear size in the principal plane of the crystal), which adversely affects its divergence and, hence, the conversion efficiency. This scheme was first experimentally realised in papers [26, 27].

In this paper, we used the improved two-pass amplifier [28] with the maximum peak power 300 kW and the average power up to 25 W. A nonlinear crystal was irradiated by a laser beam of a rectangular cross section in the focal plane, which was formed by cylindrical lenses and had a low divergence in the principal plane of the crystal. For comparison, we also performed experiments using a parallel laser beam with circular cross section.

2. Experimental

The optical scheme of the experiment is shown in Fig. 1. The design and parameters of the amplifier are described in detail in [27, 28]. The setup consisted of master oscillator (*1*), polarisation coupler (*2*), amplifying stage (*3*), beam-return unit (*4*) including phase quarter-wave plate (*5*), and plane mirror (*6*). The amplifying stage based on a GL-201 tube with a 80-cm long operating chamber and diameter of 20 mm emitted 0.51-μm and 0.578-μm pulses with a pulse repetition rate of 10 kHz. The output beam J^+ of the master oscillator with the horizontal polarisation of the electric vector (in the figure plane) entered into the active medium of amplifying stage (*3*) and then into beam-return unit (*4*). The beam J^- propagating in the opposite direction acquired orthogonal polarisation and therefore it did not interfere with the beam J^+ and was coupled out from the system by polarisation coupler (*3*) and directed to fold mirror (*7*) and then to telescopic collimator (*8*) consisting of spherical lenses. To reduce the laser radiation losses and optical aberrations during the round-trip transit of the laser beam, the optical windows of the amplifier tube were covered with the AR coating, and a special prism liquid-crystal polariser, which was developed and fabricated for CVLs in [29], was used as polarisation coupler (*2*). This polariser has a high transmission (0.985), good polarisation properties, and a high radiation resistance.

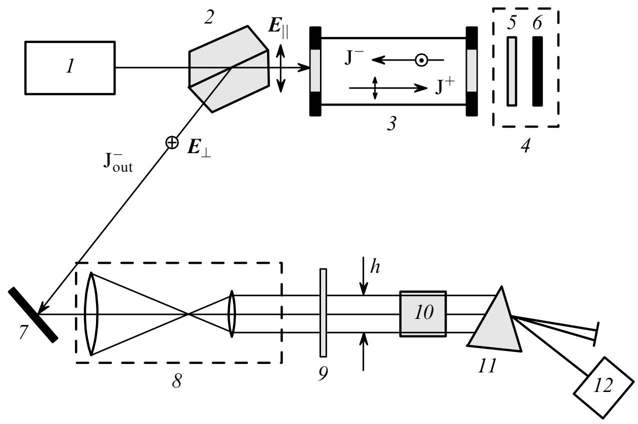

Figure 1. Scheme of the experiment: (*1*) master oscillator; (*2*) polarisation coupler; (*3*) amplifying stage; (*4*) beam-return unit; (*5*) phase quarter-wave plate; (*6*) plane mirror; (*7*) fold mirror; (*8*) telescopic collimator; (*9*) cylindrical lens; (*10*) nonlinear crystal; (*11*) quartz prism; (*12*) power meter.

In the first series of experiments, the laser beam after collimator (*8*) propagated through cylindrical lens (*9*), which compressed it only along one axis perpendicular to the figure plane (the principal plane of the crystal). The centre of the beam waist was located in the middle of crystal (*10*). The beam cross section near its waist was approximately rectangular, with the width $h \approx 5 - 9$ mm (Fig. 1) equal to the beam diameter at the output of collimator (*8*). The value of h was selected as large as possible and was limited only by the transverse size of the crystal. This was done to obtain the minimal, diffraction-limited divergence of the beam in the principal plane of the crystal, which is determined by the value of h.

In the second series of experiments, the beam diameter was reduced down to 1–1.2 mm with the help of telescopic collimator (*8*) and then this beam (with circular cross section) propagated through nonlinear crystal (*10*).

The divergence φ (the full angle) of the beams with circular and rectangular cross sections was measured by the method of calibrated apertures in air (in the principal plane of the crystal) and recalculated to the divergence φ_{cr} inside crystals taking into account the refraction law (Table 1).

We used in experiments the DKDP and BBO crystals. The DKDP crystal of diameter 10 mm and length $l = 4$ cm with the cut angle 78.5° was used for the SFG ($\lambda =$ 0.271 μm) for the two lines of the amplifier. The BBO crystal of transverse dimensions 7×7 mm and length along the laser beam $l = 10$ mm had the cut angle 48°, which allowed the SHG (0.255 μm) for the green line, the SHG (0.289 μm) for the yellow line, and SFG (0.271 μm) by

Table 1. Parameters of nonlinear crystals and laser radiation.

Crystal	Process	$\lambda/\mu m$	l/cm	θ/deg	ΔQ/mrad	h/mm	φ/mrad	φ_{cr}/mrad
DKDP	SFG	0.271	4	78.5	0.39	1.1–1.2* 9	2–4 0.3–0.4	1.3–2.6 0.2–0.26
BBO	SFG	0.271	1	46.3	0.18	1.1–1.2* 5.5	2–4 0.5–0.66	1.3–2.6 0.3–0.4
BBO	SHG	0.255	1	50.7	0.18	1.1–1.2* 5.5	2–4 0.5–0.66	1.2–2.4 0.3–0.4
BBO	SHG	0.289	1	42.5	0.26	5.5	0.5–0.66	0.3–0.4

*Note: Diameter of a laser beam with a circular cross section.

tuning the phase-matching angle. In all the above cases, the scalar oo–e phase matching was used.

The phase-matching angle θ and the angular phase-matching width ΔQ (for the specified length l of the crystal) calculated inside crystals for our experimental conditions are presented in Table 1. These parameters were calculated using the properties of the crystals presented in [30–32]. The crystal temperature was assumed equal to 40 °C.

The UV radiation was separated from laser radiation with quartz prism (*11*). The UV radiation power W_{UV} averaged over the pulse repetition rate was measured with calorimeter (*12*) and recalculated to the crystal output taking into account losses in prism (*11*). The same calorimeter was used to measure the average powers of laser radiation at the crystal input: the total power at both wavelengths (W_{g+y}), and at the yellow (W_y) and green (W_g) lines. The corresponding oscillograms $U(t)$ of individual radiation pulses at the crystal input and UV radiation pulses at the crystal output were recorded with a photocell and a broadband oscilloscope. The average optical efficiency of nonlinear conversion was defined $\eta = (W_{UV}/W)$, where one of the quantities W_{g+y}, W_y, or W_g was used as W.

3. Experimental results and discussion

Due to a special design of a two-pass amplifier, we obtained the doubled peak power of the output pulse compared to the single-pass amplifier for the same pump power (3.0–3.2 kW) of amplifying stages and equal average output powers. Two operation regimes of the amplifier were used in experiments. In the first regime, the maximum peak power from 210 to 300 kW was used, the average power was 22–25 W, and the ratio $W_y/W_g \approx 0.7$. In the second regime, the peak output power of the amplifier was ~ 190 kW, the average power was ~ 18.5 W, and the ratio $W_y/W_g \approx 1.0$.. Taking into account losses in optical paths, the value of W_{g+y} (at the crystal input) was varied from 13 to 17 W in different experiments.

Table 1 presents the measured divergence φ of the beams, which depended, of course, on the beam diameter at the output of telescopic collimator (*8*) (Fig. 1). Note that a careful adjustment of the telescopic collimator provided the compensation of the thermal-lens effect of the amplifying stage and ensured the minimal divergence equal to two–three diffraction-limited divergences corresponding to the size of h or the diameter of the beam with a circular cross section. Cylindrical lenses with different focal distances f_c produced different laser power densities in the crystal.

Figure 2 shows typical oscillograms of radiation pulses at the crystal input for the first and second operating regimes of the amplifier. One can see that the ratios of the peak powers of yellow (U_y) and green (U_g) radiation pulses in the first and second regimes are substantially different. It is clear that the second regime is more preferable for SFG.

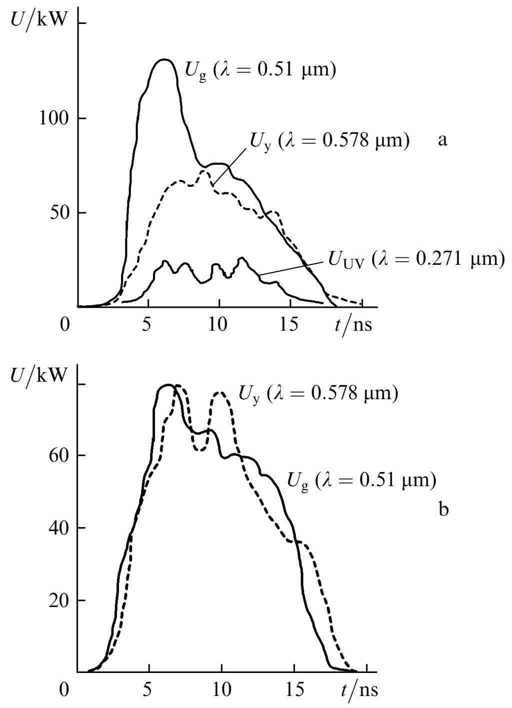

Figure 2. Oscillograms of radiation pulses at the crystal input for the first operating regime of the amplifier and $W_{g+y} = 16.8$ W (a) and for the second operating regime of the amplifier and $W_{g+y} = 15.0$ W (b).

The results of generation of UV radiation by using the beam with a rectangular cross section of width h and the beam with a circular cross section are presented in Tables 2 and 3, respectively.

The best results were obtained for the beam with a rectangular cross section both for the BBO and DKDP crystals. For the total power of the laser system equal to 18.5 W and the power at the BBO crystal input of the order of 15 W, we achieved comparatively high average powers and average optical efficiencies: 3.6 W and 24 % for SFG, 3.4 W and 44 % for SHG on the yellow line, and 2.1 W and 27 % for SHG on the green line. By using the DKDP crystal, we obtained 2.1 W of the SFG power and the 14 % efficiency. For the beam of a circular cross section, the UV radiation power and efficiency did not exceed 1.8 W and 11 %, respectively, in the case of SFG using DKDP and were ~ 1 W and 5 %–8 % for the BBO crystal.

Table 2. Parameters of UV radiation generation upon irradiation of a crystal by a laser beam with a rectangular cross section of width h in the principal plane of the crystal (second operation regime of the amplifier).

Crystal	Process	$\lambda/\mu m$	W_{g+y}/W	W_y/W_g	U_g/kW	U_y/kW	h/mm	f_c/mm	W_{UV}/W	η (%)
DKDP	SFH	0.271	14.6	0.9	80	72	9.0	400	2.1	14
DKDP	SFH	0.271	15.4	0.9	80	72	9.0	150	1.54	10
BBO	SFH	0.271	15.0	1.0	77.6	77.6	5.5	150	3.6	24
BBO	SHG	0.255	15.6	1.0	80	–	5.5	150	2.1	27
BBO	SHG	0.289	15.6	1.0	–	80	5.5	150	3.4	44

Table 3. Parameters of UV radiation generation upon irradiation of a crystal by a parallel laser beam of a circular cross section.

Crystal	Process	$\lambda/\mu m$	W_{g+y}/W	W_y/W_g	U_g/kW	U_y/kW	W_{UV}/W	η (%)
DKDP	SFH	0.271	16.8	0.8	130	70	1.8*	10.7
DKDP	SFH	0.271	14.5	0.7	120	50	1.2	8
BBO	SHG	0.255	14.8	0.7	122	51	1.0	7
BBO	SFH	0.271	13.0	0.7	110	45	0.7	5.4

*Note: Corresponds to the first operating regime of the amplifier (see Fig. 2a).

The higher UV radiation powers and optical efficiencies obtained with beams with comparatively large widths $h = 5.5 - 9$ mm can be explained by the fact that the divergence φ_{cr} of the beam of a rectangular cross section is approximately an order of magnitude lower than that of the beam with a circular cross section. Note that for a beam with a circular cross section with a high divergence and small transverse size, the SFG power is higher in the DKDP crystal (1.8 W) than in BBO. This is explained by a considerably larger angular phase-matching width ΔQ for DKDP compared to BBO (Table 1).

Although the divergence φ_{cr} of the rectangular-cross section beam in these experiments with the BBO crystal was low, it exceeded the angular phase-matching width ΔQ (see Table 1). The ratio $\varphi_{cr}/\Delta Q$ was 1.2–2.2 (for the circular-cross section beam, this ratio is approximately an order of magnitude higher). It seems that for increasing the efficiency of nonlinear conversion, it is necessary to reduce the beam divergence in the crystal to the value smaller than or equal to ΔQ by increasing the beam width h and eliminating various optical aberrations.

It is obvious that, by increasing the beam width (and, therefore, the sectional area), it is also necessary to increase the output peak power of the amplifier to maintain a sufficiently high peak power density of incident radiation, as was done in the paper.

4. Conclusions

We have demonstrated the possibility of the development of efficient UV radiation sources based on multipass copper vapour laser amplifiers with enhanced output peak pulse powers. By using a 20-W two-pass amplifier in combination with the DKDP and BBO nonlinear crystals, we obtained UV radiation powers (SHG and SFG) from 2.1 to 3.6 W. Such powers have been earlier obtained by using CVLs with output powers as high as 40–50 W. Note that we have obtained a rather high average optical efficiency of 44 % and power of 3.4 W for SHG on the 0.289-μm yellow line.

The multipass amplifier should be technically modified and the scheme of beam formation in nonlinear crystals should be further optimised.

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