Pure Appl. Opt. **5** (1996) 119–124. Printed in the UK

Frequency doubling of He–Ne laser radiation at 632.8 nm

R Stoleru†, M L Pascu and A M Dumitras
Institute of Atomic Physics, Laser Department, PO Box MG-6, Bucharest 76900, Magurele, Romania

Received 11 May 1995, in final form 14 September 1995

Abstract. Second harmonic generation (SHG) obtained using a potassium dihydrogen phosphate (KDP) nonlinear crystal from a CW He–Ne laser as a pump source, which emits at 632.8 nm, is reported. The SHG efficiency at the power of the He–Ne laser utilized (18 mW) and the fundamental beam waist available (0.1 mm), was evaluated theoretically; the same efficiency was measured experimentally obtaining a value of $(3.7–0.1)\times 10^{-7}$, in good agreement with theoretical predictions.

1. Introduction

Second harmonic generation (SHG) starting from a He–Ne laser fundamental beam emitted in the spectral range 1.15–3.39 μm has been obtained and reported previously for crystals such as $LiNbO_2$ [1, 2], $AgGaS_2$ [3] and $LiIO_3$ (intracavity SHG) [4].

In this paper, the generation of the second harmonic (SH) using the He–Ne laser beam emitted at 632.8 nm as the fundamental beam in a potassium dihydrogen phosphate (KDP) nonlinear crystal is reported.

The interest in developing CW UV laser radiation sources is due to the lack of such sources to be used in spectroscopic experiments. Reports on UV pulsed laser radiation obtained at 265 nm by two-step SHG effects from YAG:Nd lasers emitting at 1.06 μm are well known [5]. Tunable pulsed UV laser radiation sources are also reported using tunable dye laser fundamental beams and one-step SHG; in this case, an UV tunability range between 250 nm and 300 nm is commonly obtained [6], while pumping an ADP or KDP crystal.

Another reported result concerns the frequency doubling by SHG in nonlinear crystals from CW tunable dye lasers, particularly from Ar^+ pumped dye lasers [7]; the UV tunability range covered is narrower in this case than for the former situation, i.e. typically 290–310 nm.

There are, in principle, many possibilities to obtain CW UV tunable laser radiation starting from recently developed diode lasers which emit in the visible or from some other types of lasers, in this last case at relatively high costs.

Nevertheless, it seems that using a He–Ne laser beam at 632.8 nm as the fundamental beam to obtain UV laser radiation by SHG in nonlinear crystals at fixed wavelength is still convenient, because of the low costs and simple experimental arrangement. The UV beam may be used in experiments such as laser induced fluorescence and phosphorescence for complex molecules (biomolecules, pollutants), optogalvanic spectroscopy measurements and alkali metals and/or alkali metal/noble gas mixtures, etc.

† E-mail: stoleru@ifa.ro

0963-9659/96/010119+06$19.50

2. Theoretical considerations

The SHG in the reported case has been realized by index matching using a type I process in a negative uniaxial crystal, KDP [8].

KDP has been chosen since it exhibits a good transmittance (more than 85% with a crystal length of 20 mm) in the spectral range of interest, 300–700 nm [9]. In figure 1 are plotted the indices of refraction for ordinary rays and for extraordinary rays, respectively, against the wavelengths of the fundamental beam, for a KDP crystal [10]. For 632.8 nm fundamental beam wavelength, the refractive indices of the ordinary and extraordinary rays, used further, are $n_o = 1.5074$ and $n_e = 1.492$, respectively.

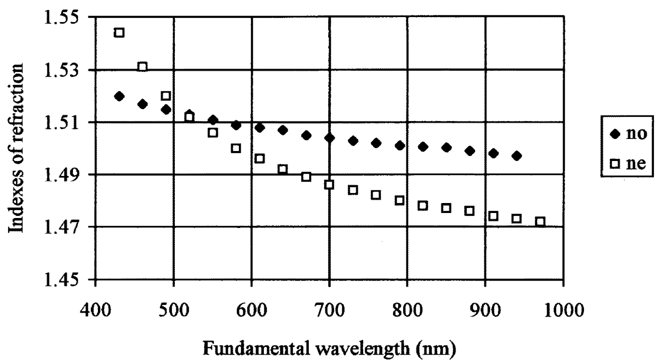

Figure 1. Refractive indices for ordinary rays and for extraordinary rays, plotted against the wavelengths of the fundamental beam.

Both index-matching (θ_m) and birefringence (ρ) angles have been calculated according to equations specified in [10]:

$$\sin^2\theta_m = \frac{(n_e^{2\omega_1})^2[(n_o^{2\omega_1})^2 - (n_o^{\omega_1})^2]}{(n_o^{\omega_1})^2[(n_o^{2\omega_1})^2 - (n_e^{2\omega_1})^2]} \tag{1}$$

where ω_1 is the angular frequency of the fundamental beam,

$$\tan\rho = \tfrac{1}{2}(n_o^{\omega_1})^2\left[\frac{1}{(n_e^{2\omega_1})^2} - \frac{1}{(n_o^{2\omega_1})^2}\right]\sin(2\theta_m). \tag{2}$$

The following values were obtained: $\theta_m = 56.1^\circ$ and $\rho = 0.0281$ rad.

The nonlinear coefficient for the KDP crystal is: $d_{36}(632.8\text{ nm}) = 7.1 \times 10^{-13}$ m V^{-1} [10].

These values allowed us to estimate theoretically the conversion efficiency of the SHG process in our particular case: continuous wave and Gaussian fundamental beam emitted by a He–Ne laser at 632.8 nm, fundamental beam power 18 mW and fundamental beam waist $w_0 = 0.1$ mm.

An important feature to improve the SH efficiency is the focalization. On the other hand, by reducing w_0, the divergence increases: so it is necessary in all cases to look for the optimal value of w_0, which gives optimization of the SH power. Taking into account the absorption for the primary and SH fields travelling across the crystal, by means of the macroscopic quantities α_1 (absorption coefficient for the fundamental beam) and α_2 (absorption coefficient for the frequency doubled radiation) respectively, the general expression of SH power P_2 is [11]:

$$P_2 = K P_1^2 \frac{\ell^2}{w_0^2} \exp(-\alpha_2 \ell) G(t, q) \tag{3}$$

where P_1 is the fundamental beam power, ℓ is the crystal length and K is a coefficient depending on d_{ooe} (the nonlinear effective coefficient) and on θ_{m}, given by:

$$K = \frac{128\pi^2 \omega_1^2}{(n_1 c^3)} d_{\text{ooe}}^2 \sin^2 \theta_{\text{m}} \tag{4}$$

where n_1 is the fundamental beam refractive index and c the light speed; G is a function which may be calculated numerically for different values of t and q, i.e. the parameters which take into account the absorption and birefringence: $t = \sqrt{2}\rho\ell/w_0$; $q = \alpha w_0/2\sqrt{2}\rho$; $\alpha = \alpha_1 - \frac{1}{2}\alpha_2$.

The full expression of G is:

$$G(t, q) = \sqrt{\frac{2}{\pi}} \int_{-\infty}^{\infty} F^2(u, t, q)\, \mathrm{d}u \tag{5}$$

with the limit condition: $G(0, 0) = 1$.

In equation (5), $F(u, t, q)$ is given by:

$$F(u, t, q) = \frac{1}{t} \int_0^{\infty} \exp(-2q\tau - (u + \tau)^2)\, \mathrm{d}\tau \tag{6}$$

where: $u = \sqrt{2}x/w_0$; $t = \sqrt{2}\rho z/w_0$; the expression given in (6) for $F(u, t, q)$ contains the current variable change from x, z in the plane perpendicular to the fundamental beam propagation direction to u and τ.

In the case of $q \Rightarrow 0$ (negligible absorption) it is easy to find the asymptotic behaviour of $G(t, q)$. It is shown [11] that if $q = 0$ and $t \gg 1$ (non-negligible birefringence) then:

$$P_2 = K P_1^2 \frac{\ell \ell_{\text{a}}}{w_0^2} \tag{7}$$

where

$$\ell_{\text{a}} = \sqrt{\pi}\, w_0/\rho \tag{8}$$

is the aperture length.

We have computed ℓ_{a} and the focal length l_{f} given by $\ell_{\text{f}} = \frac{1}{2}\pi k_1 w_0^2$ where k_1 is the wave vector of the fundamental beam. The values obtained are: $\ell_{\text{a}} = 6.3$ mm and $\ell_{\text{f}} = 155$ mm. The length of the utilized KDP crystal is $\ell = 20$ mm. These computations allow us to estimate that our experimental conditions are situated in the region where double refraction is not negligible ($\ell_{\text{a}} \ll \ell \ll \ell_{\text{f}}$). The expression of the SH power is, according to equations (7) and (8):

$$P_2 = K P_1^2 \sqrt{\pi} \frac{\ell}{w_0 \rho}. \tag{9}$$

Figure 2 shows the SH power plotted against fundamental beam power for the He–Ne laser wavelength, and a beam waist $w_0 = 0.1$ mm.

Figure 3 shows the SH beam efficiency plotted against the fundamental wavelength within the transparency range of the utilized KDP crystal.

For a crystal length of 2 cm, the theoretically computed conversion efficiency, defined as the ratio between the SH beam power and the fundamental beam power, was obtained to be $(3.0 \pm 0.1) \times 10^{-7}$.

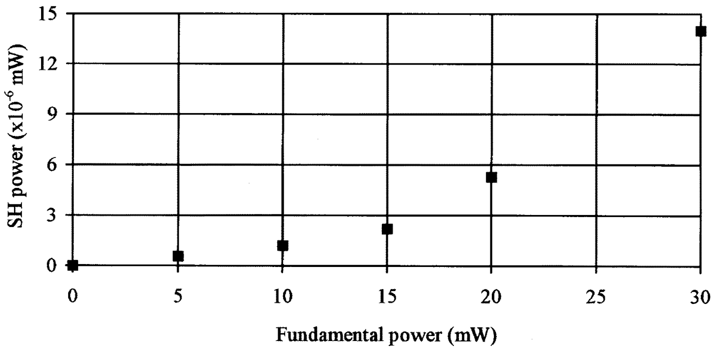

Figure 2. SH power plotted against the fundamental beam power for the He–Ne laser and the beam waist $w_0 = 0.1$ mm.

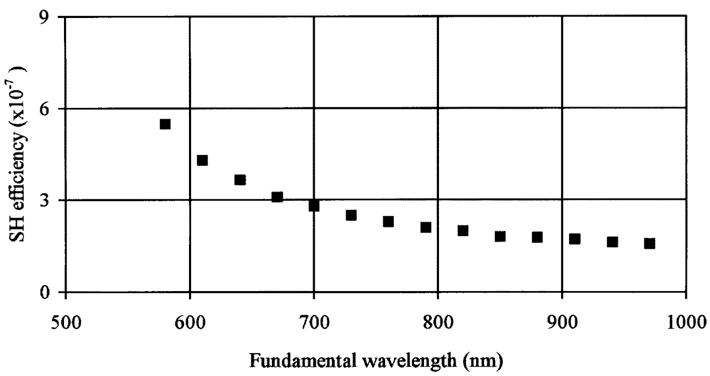

Figure 3. SH beam efficiency plotted against the fundamental wavelength.

3. Experimental set-up and results

The experimental set-up was conceived to allow carrying out SHG by means of a fundamental beam properly focused and linearly polarized, so that the propagation directions of the fields in the crystal correspond to that of index matching. At the same time, the experimental arrangement was conceived to measure small intensity UV laser radiation.

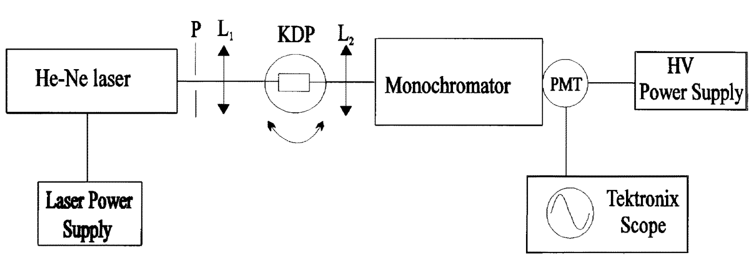

Figure 4. The experimental set-up.

The experimental set-up (figure 4) contained a CW homemade LGA-50 He–Ne laser as the fundamental beam source, which operated in multimode emission, with an output power level of 25 mW. A pinhole P was used to allow working with TEM_{00} laser beam mode structure by spatial filtering; the beam is vertically polarized. The fundamental beam power used for the SHG experiment was, after filtering, 18 mW. In order to achieve maximum available conversion efficiency and easily observable UV outputs, it was convenient to tightly focus the fundamental beam, with a $f = 10$ cm lens, into the homemade KDP crystal; this allowed us to work with a fundamental beam waist of about 0.1 mm, which is the value considered in the theoretical computations. An SPM2 monochromator (Zeiss–Jena) was used to select the SH beam at 316.4 nm originating from the fundamental beam at 632.8 nm. The UV radiation was detected using an 1P28 Hamamatsu photomultiplier. Given the difference between the wavelengths of interest ($\lambda_{He–Ne} = 632.8$ nm and $\lambda_{SHG} = 316.4$ nm) we had to use two different gratings. The first one works in the range 360–720 nm and the second between 200 nm and 610 nm. The signals monitored on a 7904 Tektronix scope had to be corrected in order to take into account the dispersion efficiency of each grating for the two orthogonally polarized beams. To do it, we used a satellite line of the He–Ne laser, emitted at 383 nm, which is common for both gratings. We measured the laser beam powers at 383 nm when analysed with both gratings and a correction factor of 1.4 resulted, i.e. we had to multiply by 1.4 the measured signal at 316.4 nm and to use the measured signal, as obtained, at 632.8 nm. Another correction was necessary since the relative sensitivities of the photomultiplier in the UV and the visible were different [12]. For the UV wavelength (316.4 nm) the mentioned relative sensitivity is 90% whereas for the visible (632.8 nm) it is 9%; consequently a multiplication by a factor of 10 was made on the He–Ne laser fundamental beam measured signal.

The reported He–Ne laser fundamental beam power was measured using a homemade metrological powermeter dedicated for He–Ne laser beam monitoring.

From the performed experimental measurements we obtained a conversion efficiency for the SHG process of $(3.7–0.1)\times10^{-7}$.

4. Conclusions

The reported conversion efficiency measured experimentally is in good agreement with theoretical computations. Both efficiencies were nevertheless affected by errors. The experimental errors, for instance, are related to the fact that the He–Ne laser fundamental beam is vertically polarized, whereas the UV generated radiation is horizontally polarized (type I frequency matching process). In this case, the diffraction efficiency of the grating used for the UV beam monitoring is diminished by 10%.

The computed value of the conversion efficiency is affected by the cumulated errors on the θ_m, ρ and d_{ooe} calculated values.

At the same time, one may appreciate that the obtained result shows the availability of CW UV laser radiation obtained from a cheap and common He–Ne laser; the UV radiation has, for the moment, a major drawback, mainly the low power level, which is in the range of nW, as results from the measured conversion efficiency.

For spectroscopic measurements, an increased power level of the UV radiation is needed, which may be obtained along the following experimental lines.

(a) Lowering the beam waist through the crystal several times, which may be responsible for a corresponding UV radiation power increase (see equation (9)), outside the cavity mounted crystal. The computed value of the waist that optimizes the SH power is 0.03 mm instead of the 0.1 mm available in our experiment.

(b) Increasing the He–Ne fundamental beam power, which may be followed by an increase in the power of the UV radiation by a factor of 3–5, outside the cavity mounted crystal.

(c) Introducing the nonlinear crystal within the Fabry–Perot type cavity of the He–Ne laser; in this case, a working regime of intracavity generation of second harmonic is obtained.

If a proper cavity length is chosen and spatial filtering is performed, the TEM_{00} oscillating mode of the He–Ne laser radiation may be used.

According to literature reports [13], in optimal conditions (maximum reflection coefficients of the mirrors in the visible and as small as possible in the UV) the efficiency of second harmonic generation for CW lasers is in the range 5%–10%, so that an improvement in the UV radiation power of several orders of magnitude is to be expected.

Acknowledgments

The authors acknowledge the assistance in carrying out this work of Mr S Hogiu and Mr N Moise. They are particularly indebted to Dr M Ristici for making available the He–Ne laser.

References

[1] Antonov E N, Koloshnikov V G and Nikogosyan D N 1974 *Opt. Spectrosk.* **36** 768
[2] Kushida T, Tanaka Y, Ojima M and Nakazaki Y 1975 *Japan. J. Appl. Phys.* **14** 1097
[3] Badikov V V, Pivovarov O N, Skokov Yu V, Skrebneva O V and Trotsenko N K 1975 *Kvant. Elektron.* **2** 618 (Engl. transl. 1975 *Sov. J. Quantum Electron.* **5** 350)
[4] Kaczmarek F and Jendrzejczak A 1983 *Opt. Quantum Electron.* **15** 187
[5] Yariv A 1971 *Introduction to Optical Electronics* (New York: Holt, Rinehart and Winston) p 195
[6] Pascu M L *et al* 1988 *Theoretical and Experimental Studies on Tunable Dye Lasers* (SPIE **1033**) (New York: SPIE) pp 122–9
[7] Labuda E F and Johnson A M 1967 *IEEE J. Quantum Electron.* **3** 164
[8] Castelleto S, Godone A, Novero C and Rahimullah K 1993 Second harmonic generation with semiconductor lasers in the spectral region of 400 nm Private Communication
[9] Lee Smith W 1975 *Appl. Opt.* **16** 1798
[10] Dmitriev V G and Gurzadyan G G 1991 *Handbook of Nonlinear Optical Crystals* (Berlin: Springer)
[11] Boyd G D, Ashkin A, Djiedzic J M and Kleinmann D A 1965 *Phys. Rev.* **137** A1305
[12] RCA 1P28 Data sheet 1981
[13] Dmitriev V and Tarassov L 1987 *Optique Non Linéaire Appliquée* (Moscow: Mir)