85% efficiency for cw frequency doubling from 1.08 to 0.54 μm

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Conversion efficiency of 85% has been achieved in cw second-harmonic generation from 1.08 to 0.54 μm with a potassium titanyl phosphate crystal inside an external ring cavity. An absolute comparison between the experimental data and a simple theory is made and shows good agreement.

Because of the nonlinear nature of the process and the generally small susceptibilities involved, harmonic generation with high conversion efficiency is most often realized only for pulsed radiation with high intensity for a short period.[1] By contrast, the intensity available for cw operation is usually limited to much smaller values than for pulsed operation, with a concomitant reduction in conversion efficiency. A possible remedy to this circumstance that was recognized long ago is to employ a high-finesse resonator in either a passive[2] or active[3] configuration to enhance the circulating power and hence the overall conversion efficiency. While the quest for efficient conversion in a cw setting might at first sight seem to be best pursued with crystals with large nonlinear coefficients, such as lithium niobate and potassium niobate,[4,5] unfortunately the advantages of large nonlinear coefficient are often mitigated by deleterious effects such as linear and nonlinear absorption and subtle intensity-dependent changes in refractive index, which although small can quickly degrade the performance of a high-finesse cavity. On the other hand, crystals with greater transparency, with fewer thermal problems, and with a higher damage threshold such as the newly developed lithium triborate usually have much smaller nonlinear coefficients and therefore require correspondingly higher powers for the fundamental wave.[6]

For many years, potassium titanyl phosphate (KTP) has been a well-known crystal for frequency doubling of Nd:YAG lasers (1.064 μm).[7] The large nonlinear coefficient of KTP together with the wide temperature width, large acceptance angle, and extremely low absorption loss make KTP a promising candidate for high conversion efficiency. However, because type II critical angle tuning is the only possible phase-matching scheme at 1.064 μm, the performance of KTP inside a high-finesse resonator is significantly degraded owing to the walk-off of ordinary and extraordinary beams within the crystal. To circumvent this difficulty, recently Garmash *et al.*[8] reported type II 90° noncritical phase matching in an a-cut KTP crystal at 1.08 μm, thus suggesting new possibilities for better performance of KTP in intracavity cw frequency doubling. Following this lead, we report in this Letter experiments for frequency doubling from 1.08 to 0.54 μm with a single KTP crystal inside an external ring cavity of extremely low passive loss. Our measurements are carried out over a range of input fundamental powers up to 700 mW, and a cw conversion efficiency of 85% is achieved. The experimental results are in good absolute agreement with a simple theory, which suggests that efficiencies greater than 90% should be obtainable in our current system once the focusing geometry is optimized.

As illustrated in Fig. 1, our frequency-doubling experiment consists of a folded ring cavity of total length 38 cm with two curved mirrors (M1, M2) of 10-cm radius of curvature and two flat mirrors (M3, M4). A KTP crystal is placed between the two curved mirrors at the position of the collimated waist ($\omega_0 \cong 60$ μm), with a small folding angle (<6°) used in the arrangement to reduce both astigmatism and polarization imbalance. Three of the four mirrors have high reflectivity at 1.08 μm ($R > 99.98\%$) and high transmissivity at 0.54 μm ($T \sim 94$–96%). Infrared excitation power P_1 is coupled into the cavity through the remaining flat mirror M4 with reflectivity 97% at 1.08 μm. The fundamental wave in our experiment comes from a single-frequency, TEM$_{00}$-mode Nd:YAlO$_3$ laser operating at 1.08 μm, with the frequency of the laser locked to a stable external cavity and with 700 mW of infrared power available to pump the doubling cavity. The length of the doubling cavity is servo controlled with a standard locking system[9] to obtain coincidence between the fundamental input frequency and the frequency of a TEM$_{00}$ longitudinal-mode resonance for the ordinary polarization. The harmonic light generated in the KTP crystal is transmitted principally through the curved mirror M2 (with transmission at 0.54 μm of 94%) as shown in Fig. 1.

The nonlinear medium for frequency doubling is a 3 mm × 3 mm × 10 mm flux-grown, a-cut KTP crystal with two faces antireflection coated for low loss at both fundamental and harmonic wavelengths. Type II noncritical 90° phase matching at 1.08 μm $[2n_o(2\omega) = n_e(\omega) + n_o(\omega)]$ is achieved near 63°C with a temperature width of 30°C for our crystal (note that this phase-matching temperature is significantly different from the temperature of 153°C reported in Ref. 8).[10] Because type II phase matching requires both ordinary (o) and extraordinary (e) beams for the generation of harmonic light, the in-

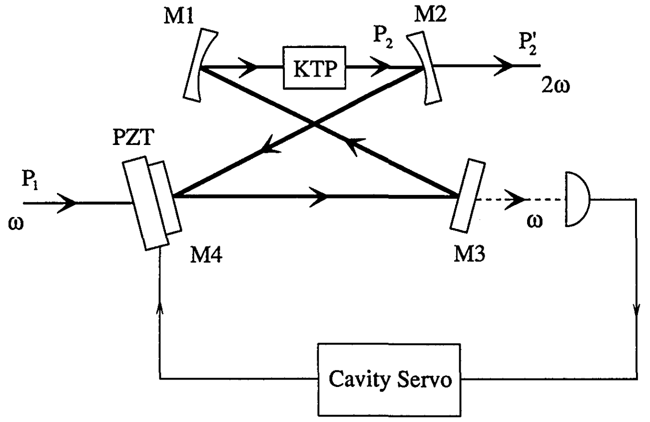

Fig. 1. Schematic of the experimental setup. PZT, piezoelectric transducer.

put infrared light is polarized at 45° with respect to the *b* or *c* axis of the crystal. The generated green light is polarized along the *b* axis. One problem encountered in type II doubling in a resonant cavity is that the indices of refraction are different for the ordinary and extraordinary beams circulating through the KTP crystal, so that longitudinal modes of the doubling cavity corresponding to the *e* and *o* polarizations are generally not simultaneously resonant for a given geometrical length of the cavity. However, the indices of refraction for the *e* and *o* beams have different temperature dependencies so that temperature tuning over a range of approximately 10°C within the wide phase-matching width suffices to bring longitudinal modes for *e* and *o* beams simultaneously into resonance. The temperature tolerance for maintaining overlapping resonance is approximately 20 mK for our arrangement. In fact, during the experiment, we actively stabilize the temperature of the crystal to a few millikelvins, so that the generated green power is stable to a few percent over 5–10 min.

For measurements of power a calibrated thermal power meter (Coherent Model 210) is employed for both input fundamental (P_1) and output harmonic (P_2) waves, except at low input power (<20 mW), where a calibrated photodiode is used. From these measured powers, we compute the total conversion efficiency $\eta = P_2/P_1 = (P_2'/0.94)/P_1$, where the factor 0.94 is the measured transmission at 0.54 μm of the cavity mirror M2. In Fig. 2 we display the results of our measurements over a range of fundamental input powers from 5 to 700 mW, with Fig. 2(a) giving the dependence of the harmonic output P_2 on infrared input P_1 and Fig. 2(b) displaying the dependence of the conversion efficiency η on P_1. In particular, for an infrared input power $P_1 = (700 \pm 10)$ mW, a green output power $P_2' = (560 \pm 5)$ mW is obtained, representing a directly measured conversion efficiency of 80%. After considering the 94% transmission of mirror M2, we obtain an overall conversion efficiency of (85 ± 1)% for frequency doubling from 1.08 to 0.54 μm. We stress that the correction for the 6% Fresnel loss at the curved mirror is the only adjustment made to the data.

For frequency doubling in either external passive cavities or active laser cavities, thermal effects driven by absorption of fundamental and/or harmonic radiation can have an adverse impact on the harmonic conversion.[5] Fortunately we have found that for our range of operating power, the KTP crystal shows only slight thermal effects. In particular, the temperature of the crystal rises as the fundamental wave builds up in the cavity, with the magnitude of the temperature rise varying by as much as 0.5°C for an infrared input power of 700 mW (note that $P_1 = 700$ mW corresponds to approximately 30 W of circulating infrared power). This small temperature rise seems to be associated with no major deleterious thermal effects, as we can see from the good agreement in Fig. 2 between the experimental data and the theoretical curves. These theoretical results are derived from a simple analysis of second-harmonic generation in an external cavity.[2,5] If we denote the overall conversion efficiency by $\eta \equiv P_2/P_1$ with total generated harmonic power P_2 and input fundamental power P_1, it is straightforward to show that

$$\sqrt{\eta} = \frac{4T\sqrt{E_{\mathrm{NL}}P_1}}{[2 - \sqrt{1-T}(2 - L - \sqrt{\eta E_{\mathrm{NL}} P_1})]^2}, \qquad (1)$$

where T is the transmission coefficient of the infrared input coupler (M4 in Fig. 1), L is the total infrared round-trip loss in the cavity excluding T, and E_{NL} is the single-pass nonlinear conversion efficiency without the cavity ($E_{\mathrm{NL}} = P_2/P_1^2$ for $T = 1$, $L = 0$). For our experiment E_{NL} was determined by

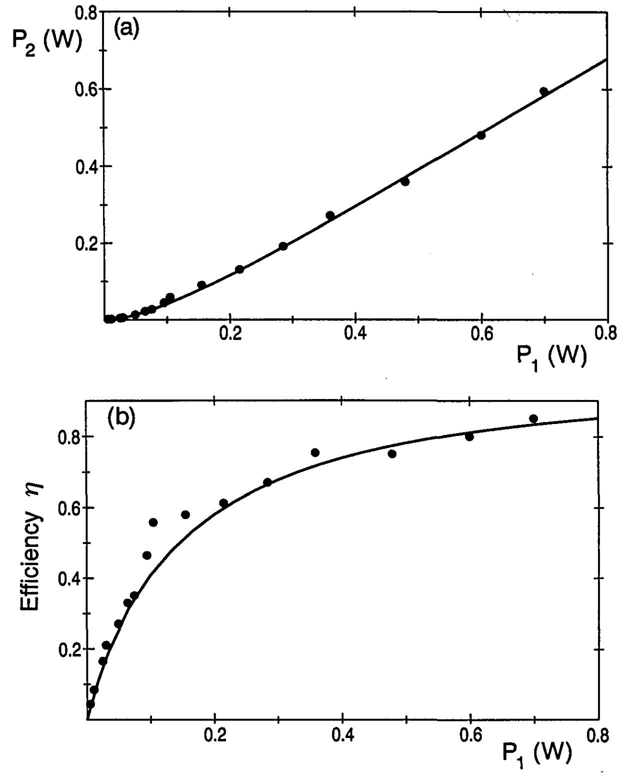

Fig. 2. (a) Second-harmonic power P_2 as a function of input infrared power P_1, (b) conversion efficiency η as a function of input infrared power P_1. The solid curves are derived from Eq. (1) with the measured values $E_{\mathrm{NL}} = 6.3 \times 10^{-4}\ \mathrm{W}^{-1}$, $L = 0.32\%$, and $T = 3\%$.

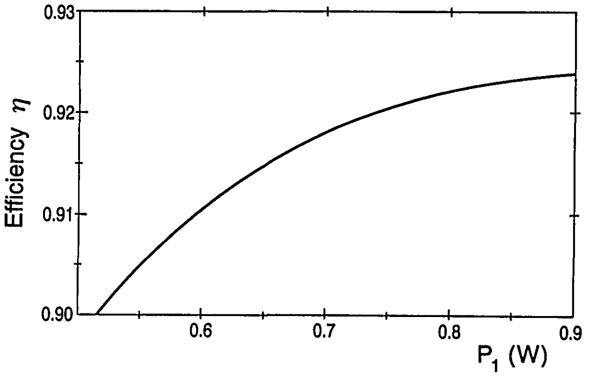

Fig. 3. Theoretical conversion efficiency η as a function of input infrared power P_1 with $E_{\mathrm{NL}} = 2 \times 10^{-3}$ W^{-1}, $L = 0.32\%$, and $T = 3.9\%$.

removing the input coupler (M4) and measuring the generated green light, which thus preserved the focusing geometry used in the actual resonant cavity. The result for the single-pass efficiency is $E_{\mathrm{NL}} = 6.3 \times 10^{-4}$ W^{-1}. One important aspect of our experiment is that the total internal loss L is quite small because of the extremely low losses of the cavity mirrors and the good quality of the KTP crystal. We determined that $L = 0.32\%$ from measurements of the finesse of the doubling cavity, with the principal contribution coming from reflection losses at the surfaces of the KTP crystal. The transmission T was directly measured to be 3%, which is close to the optimized value of 2.3% for 700 mW of infrared input power [Eq. (2) of Ref. 5]. Knowledge of T, L, and E_{NL} allows us to calculate the expected theoretical efficiency η for second-harmonic generation from Eq. (1) together with the expected power $P_2 = \eta P_1$, with the results shown in Fig. 2 (solid curves) together with the experimental data. Note that there is no fitting of the theory to the data; the comparison is in absolute terms without adjustable parameters.

As for the possible improvement of these results, we note that the optimum waist for second-harmonic generation for our 10-mm KTP crystal is approximately 20 μm,[11] which is considerably smaller than the actual waist of 60 μm in our experiment. This lack of optimum focusing is reflected in the fact that our measured value of E_{NL} is approximately three times smaller than the best value of $E_{\mathrm{NL}} \cong 2 \times 10^{-3}$ W^{-1} that we infer for this crystal from separate measurements with a longer crystal in a geometry free from the constraints imposed by the cavity. We are currently constructing a new cavity with curved mirrors (M1, M2) of radii of curvature of 5 cm, so that a waist of approximately 20 μm can be obtained in a regime of broad stability for the cavity. We should then be able to achieve $E_{\mathrm{NL}} = 2 \times 10^{-3}$ W^{-1}, which with the loss $L = 0.32\%$ (as demonstrated already in the current cavity) and with the optimized value $T = 3.9\%$ for $P_1 = 700$ mW [Eq. (2) in Ref. 5] should lead to conversion efficiency greater than 90%, as illustrated in Fig. 3.

In conclusion, we have demonstrated an efficient avenue for cw frequency doubling. Conversion efficiency of 85% has been achieved by using a KTP crystal cut for type II noncritical phase matching near 1.08 μm. Although we have employed a conventional discharge-pumped cw neodymium laser, a doubling cavity with capabilities such as ours should have broad application as an element in a system for frequency conversion from a semiconductor diode laser. There are as well a number of important problems in quantum optics for which a system with such high conversion efficiency can be gainfully employed, including operation in a conjugate mode as an efficient nondegenerate optical parametric oscillator.

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10. This difference may be due to shifts in the phase-matching temperature associated with focusing geometry (see Ref. 11). Such shifts are observed in our study for different cavity beam waists with a given crystal.
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