Broadly tunable ultraviolet light generation in a compact MgO-doped periodically-poled stoichiometric lithium tantalate optical parametric oscillator with a high-*Q* cavity

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A compact tunable UV laser source based on intracavity sum frequency generation in a MgO-doped periodically-poled stoichiometric lithium tantalate optical parametric oscillator is reported. UV output at an ~1 mW level of power over the range of 364 to 378 nm with a bandwidth of 0.5 nm was obtained with a crystal that has just one periodically-poled grating period. The full tuning range can be as much as ~68 nm, from 324 to 392 nm by varying the crystal temperature from room temperature to 250 °C. This will cover nearly the entire UVA range. © 2008 Optical Society of America

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Tunable solid-state lasers are important sources of radiation in many analytical instruments. Compact semiconductor lasers and laser diodes at the milliwatt level with output wavelengths that range from the mid-IR to the deep blue are commonly used in spectrophotometers for absorption measurements and for inducing fluorescence in gaseous molecules and in liquids. [1] However, the shortest wavelength that is easily accessed is ~390 nm, provided by GaN laser diodes. Wavelengths shorter than ~390 nm are not readily available. Meanwhile, the UVA region of radiation from 320 to 400 nm is extremely important because many molecules interact with light in this wavelength region to undergo chemical reactions. Consequently there are numerous potential applications, including UV curing/printing/coating, scientific analysis, phototherapy, air and water purification, crime investigation, and banknote inspection [2].

Generally, ~1 mW of power is sufficient for many of these applications but, depending on the application and the molecule involved, the UV wavelength suitable for each application could be different. So, it is desirable to have sources that can emit over a broad UV range with milliwatt or higher output power. While incoherent sources, such as UV lamps, are being used, it is recognized that coherent sources with their directed output beams are more effective and efficient for many of these applications. Yet most existing UV lasers emit at a single wavelength so that several different lasers are needed to cover the entire UVA region.

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Compact sources of tunable UV radiation have also been reported using the nonlinear optical techniques of quasi-phase-matched (QPM) second-harmonic generation or sum mixing [3,4]. However, the UV power obtained was mostly in the few microwatts range and the tunability was extremely limited [5–8]. Scaling up the power of these QPM devices was restricted by the difficulty in fabricating large enough crystals with a small domain period that is required for QPM in the UV and by the rapidly increasing linear and nonlinear absorption losses in the common periodically-poled material of lithium niobate and KTP. It is, therefore, interesting to develop a UV source that has good power and that encompasses this entire region in a single package.

We describe an approach that employs *non-phase-matched* sum frequency mixing as a means of UV generation. The idea is to take advantage of the wavelength insensitivity of non-phase-matched generation to achieve broad tunability while employing the large nonlinearity of periodically-poled crystals to maintain a respectable conversion efficiency. Stoichiometric lithium tantalate (SLT) has a UV cutoff wavelength at ~260 nm and nearly nonexistent photorefractive damage at 532 nm [9]. Absorption loss in the UVA region is not severe. Thus, by using SLT as the nonlinear optical material, it is possible to achieve broadly tunable UV radiation and to demonstrate output power at the milliwatt level.

Figure 1 shows the calculated efficiency of frequency summing 532 nm with near-IR light from 1100 to 1400 nm in periodically-poled SLT. The domain period is 8 μm. The 532 nm input intensity is 44 MW/cm^2. The effective nonlinearity d_{eff} is estimated from the published first-order QPM d_{eff} for SLT of 10 pm/V [10] and corrected for use in the UV region according to Miller's rule [11]. The value comes to approximately 4.1 pm/V for third-order QPM.

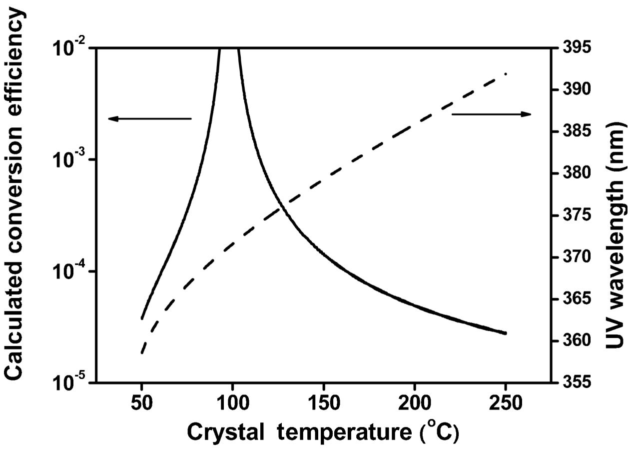

Fig. 1. Simulated conversion efficiency from the idler to the UV for third-order SFG as a function of the temperature of PPMgSLT. The domain period is 8 μm. Crystal length is 20 mm and intensity at 532 nm is 44 MW/cm^2. The corresponding UV wavelength is displayed on the right axis.

While UV generation in this crystal can employ either a first- or third-order QPM process, calculation shows that maximum conversion in this region occurs at 371 nm that corresponds to sum mixing of 532 and 1226 nm at 98 °C based on the third-order QPM process. Calculation further shows that the conversion in this region is dominated by the third-order process. The contribution from the first-order process is less than 10% of that from the third order. In Fig. 1 the solid curve has a functional form of $1/(\Delta k)^2$ and represents the conversion efficiency of a third-order QPM sum frequency generation (SFG) process that is given in mks units by

$$\frac{P_3}{P_1} = \frac{8\pi^2 d_{\mathrm{eff}}^2 L^2 P_2}{\varepsilon_0 c n_1 n_2 n_3 \lambda_3^2 A} \mathrm{sin\,c}^2(|\Delta k| L/2), \tag{1}$$

where P_i is the peak power, n_i is the refractive index, d_{eff} is the nonlinear coefficient given above, $\Delta k = k_3 - (k_1 + k_2) - 2\pi/\Lambda$, Λ is the domain period, L is the crystal length, λ_3 is the UV wavelength, A is the beam area, $\varepsilon_0 = 8.854 \times 10^{-12}$, and the indices 1, 2, and 3 represent the near-IR, 532 nm, and the UV, respectively. In this region momentum matching, $\Delta k = 0$ is at 371 nm. As can be seen, the efficiency falls off by only a factor of 10 over a spectral range of nearly 2000 cm^{-1} from 373 to 385 nm when being off from QPM and $\Delta k \neq 0$. Excluding the region from 369 to 373 nm, where the efficiency is dominated by third-order QPM centered at 371 nm, one can obtain from 358 to 385 nm with a variation of less than a factor of 10 in the UV power. The near-IR wavelength required to cover this UV range is from 1100 to 1400 nm. This near-IR range falls within the range of idler wavelengths that can be generated from a 532 nm pumped OPO that uses the same 8 μm grating period crystal [12]. In addition to the idler, sum mixing of the signal output of this OPO with 532 nm is also possible. This mixing gives tunable UV from 324 to 355 nm so that a single QPM OPO with simultaneous non-phase-matched sum generation (NPM SFG) in the same crystal can provide a total tuning range from 324 to 392 nm (a span of 68 nm) simply by changing the OPO crystal temperature from near room temperature to 250 °C. Since both the OPO and the NPM SFG employ the same crystal, this broadly tunable UV device will be compact and simple to operate.

Operation of a multikilohertz 532 nm pumped OPO using a MgO-doped periodically-poled SLT (PPMgSLT) crystal has previously been reported [12]. A tuning range from 855 to 1410 nm was achieved with a single period when operating the crystal from 30 to 200 °C. By allowing the resonating field inside this OPO cavity to build up to a high value with a low-loss (high-Q) cavity, the SFG conversion efficiency can be enhanced, permitting the generation of intense UV output throughout the tuning range of the OPO. As will be shown below, intracavity NPM SFG, therefore, is a viable approach to

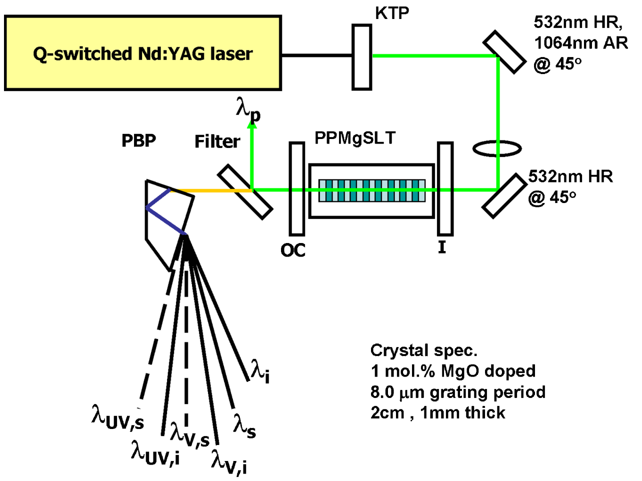

Fig. 2. (Color online) Schematic of experimental setup: OC, output coupler; I, input mirror; PBP, Pellin–Broca prism. Subscripts *p*, *i*, *s*, V, and UV stand for pump, idler, signal, visible, and UV.

generating high power broadly tunable UV in a compact setting.

Figure 2 is a schematic of the experimental setup. The output from a diode-laser-pumped *Q*-switched Nd:YAG laser was converted to the second harmonic in a type II KTP crystal, yielding 1.07 W of 532 nm radiation at a 3 kHz repetition rate. The pulse duration was 19 ns. The TEM$_{oo}$ laser beam was focused at the center of the PPMgSLT crystal to a Gaussian waist of 165 μm and a maximum intensity of 44 MW/cm^2. To reach a high resonating power inside the OPO cavity, a short, semiconfocal cavity of length 28 mm was used. A flat input mirror and a concave output coupler with a radius curvature of 5 cm formed the cavity that resonates in the idler wavelength range. The resulting optical cavity waist was about 90 μm. Both the input and output mirrors were coated for high transmission at 532 nm ($T = 95\%$) and from 350 to 390 nm ($T = 86 \pm 3\%$), and highly reflecting between 1120 and 1300 nm ($R > 99\%$). The reflectance decreases to 95% at 1108 and 1318 nm. The transmission throughout the signal wavelengths was >50% to ensure singly resonant operation at the idler wavelength. With this set of mirrors, high-*Q* (loss <5%) intracavity sum mixing of the pump and the idler would produce a UV wavelength from 361 to 378 nm.

The PPMgSLT crystal was 20 mm long and 1.0 mm thick with a 1.0 mol.% MgO doping. It was grown by the double-crucible Czochralski method and poled by a multipulse electric field of 1.4 kV/mm [9] to provide a nearly uniform domain period of 8.0 μm. The end faces of the crystal were slightly wedged and antireflection coated with $R \sim 1\%$ at 532 nm and $R < 2\%$ from 900 to 1300 nm to minimize losses inside the cavity. When operating the crystal from 50 to 130 °C, UV light from 364 to 378 nm was observed. The total UV output power (combined forward and backward directions, backward light resulting from residual reflection on the crystal and mirror surfaces) showed a peak at 369.6 nm and 77.2 °C that reached 4.7 mW when the average input pump power was 1 W (Fig. 3). This peak is the location of simultaneous OPO and third-order QPM SFG. The difference in the measured QPM UV wavelength and that from the calculation shown in Fig. 1 was a direct consequence of insufficient accuracy in the dispersion of SLT in the UV. The measured temperature width of this peak is ~3.8 °C. Taking into account that the idler bandwidth is ~4 nm, the calculated temperature width is ~3.5 °C, which is in close agreement with the measured value.

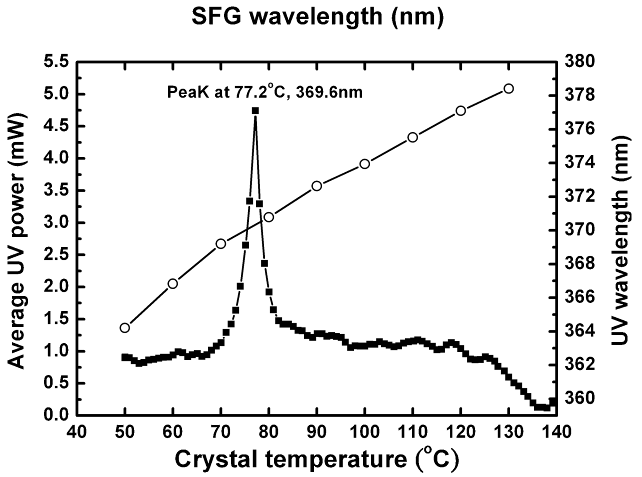

Fig. 3. Generated UV power (squares) and the corresponding tuning curve (circles) as a function of the crystal temperature. The range of wavelength covered is determined by the coating of the high-reflecting cavity mirrors.

The UV power dropped off only gradually when tuning away from this QPM location, thus resulting in a broad tuning range, as predicted in Fig. 1. For the entire range studied from 364 to 378 nm, the UV power was at the ~1 mW level. The tuning range measured was dictated by the wavelength range of high reflectivity (>99%) of the cavity mirrors. A broader range can be achieved with mirrors coated for high reflection for the full range that can be

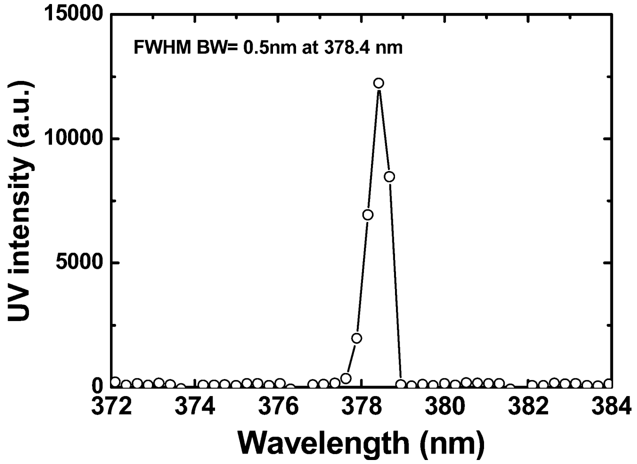

Fig. 4. Typical spectrum of the generated UV output as determined by an Ocean Optics grating spectrometer. Spectral resolution is 0.1 nm.

reached when the crystal temperature is changed. The UV bandwidth was approximately 0.5 nm, measured at full input power and with an Ocean Optic spectrometer with a resolution of 0.1 nm. This width is in agreement with expectations based on the IR bandwidth of 4 nm. Figure 4 is an example of such a UV spectrum.

The UV output at 86 °C versus 532 nm power is shown in Fig. 5. Since the OPO output has a linear dependence on the pump power when operating well above threshold [12], the measured power scaling law of 1.83 is in close agreement with the theoretical scaling law of 2 for the cascaded OPO/NPM SFG process.

The intracavity idler power is estimated to be 5.7 W, based on the measured idler output power of ~5 mW and an output-mirror reflectance of 99.915%. The estimated intracavity IR intensity is 250–300 MW/cm^2, which is a factor of 2 below the damage threshold of uncoated SLT at 1064 nm [10] so that safe long-term operation is ensured. The intracavity idler power gives an estimated NPM SFG power of ~0.1 mW using the calculated data shown in Fig. 1 and by taking into account a 50% depletion of the pump power inside the OPO cavity. This calculated UV power is about a factor of 10 less than the value measured in the experiment. Yet it is not surprising that the measured output is higher. The reason for it is likely to be similar to an observation that has been reported for non-phase-matched SHG of 1064 nm in a PPLN OPO [13] in which the authors reported that the SHG conversion efficiency reached by about 1000 times higher than their estimate.

In addition to the UV output that resulted from mixing with the idler, an intense UV peak, as shown in Fig. 6 was observed. This peak at 333.3 nm and 138.4 °C should be the result of a fifth-order QPM process of mixing the signal output with the pump. It has a lower power than the third-order QPM peak at 371 nm because the fifth-order process has a smaller d_{eff} and the signal power is weaker since it is nonresonant. The measured temperature bandwidth of 1.2 °C for this peak is dictated by the bandwidth of the signal, as is for the case of the third-order peak discussed above.

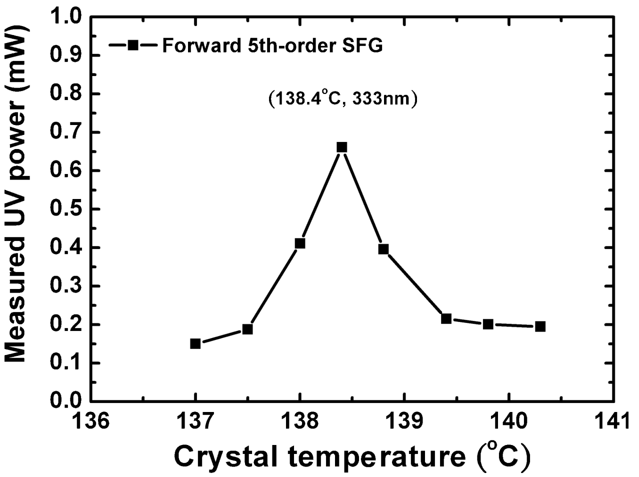

Fig. 6. UV power near 333 nm generated by fifth-order QPM SFG of the pump and the signal around 893 nm.

UV generation has also been obtained when resonating the signal rather than the idler in the OPO. By changing the OPO cavity mirrors to a pair that are highly reflecting in the signal wavelength range, we measured UV output from 329 to 348 nm for the temperature range from 40 to 190 °C, as shown in Fig. 7. We did not measure the UV power for this range because the mirrors we used have poor transmission in the UV. However, our measurements show that a broad UV tuning range of 39 nm (from 329 to 378 nm) has been achieved from the OPO with a crystal that has just one grating period so that the entire UVA range can readily be covered by operating the crystal from 30 to 250 °C. With properly coated mirrors, we can predict from the results with the idler that UV near the milliwatt level can be obtained for this range, as well.

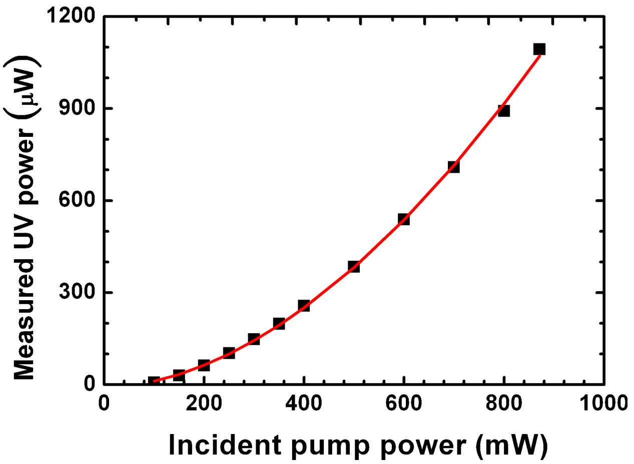

Fig. 5. (Color online) Generated UV power as a function of the incident power at 532 nm. Squares are measured values. The solid curve has a 1.83 power and is a fit to the data.

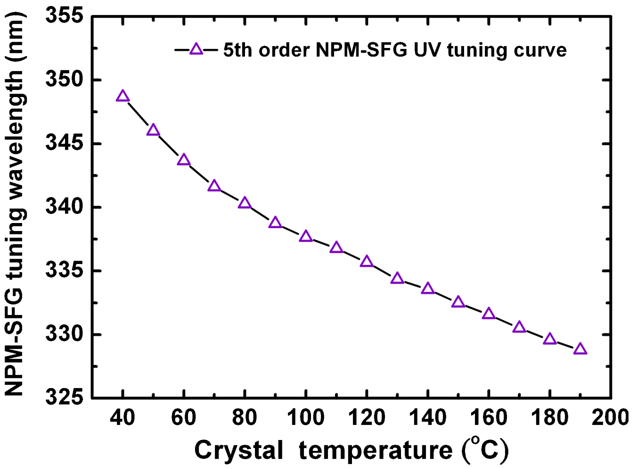

Fig. 7. (Color online) Wavelength of UV output generated by non-phase-matched fifth-order mixing of 532 nm with the signal power in the OPO as a function of crystal temperature.

To summarize, we have described a compact multikilohertz-nanosecond tunable UV light source based on intracavity SFG in a PPMgSLT OPO. UV output from 364 to 378 nm at the $\sim$1 mW level of power with a bandwidth of 0.5 nm was obtained with a crystal that had just one periodically-poled grating period. UV wavelengths outside the range reported here can be obtained by extending the operating temperature to above 250 °C to reach a full tuning range of as much as $\sim$68 nm ($5350\,\mathrm{cm}^{-1}$) from a single simple device. This type of compact and broadly tunable UV light source will be suitable for selective excitation and photochemistry of molecules, such as various analytes in liquids, and in solid-state and biological systems.

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