EFFICIENT GENERATION OF CONTINOUSLY TUNABLE ULTRAVIOLET RADIATION BY INTRA-CAVITY SUM FREQUENCY MIXING IN PULSED OPTICAL PARAMETRIC OSCILLATORS

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Abstract

Continuously tunable UV radiation was efficiently generated by intra-cavity mixing either the signal or the idler wave of a nanosecond KTP-OPO with the second or third harmonic of an injection-seeded Q-switched Nd:YAG pump laser. Starting from a pump energy of 410mJ at 1064nm a maximum energy of 19.5mJ/pulse at 320nm was achieved. This corresponds to a conversion efficiency of 9.5% from the second harmonic, or 4.8% from the fundamental of the Nd:YAG laser to the UV. The investigated set-ups could be tuned in the range of 234-325nm. The possible theoretical tuning range is as broad as 225-350nm. The divergence of the UV-beam was less than 0.7mrad. The pulse width was 5-6ns and the energy stability was 1% rms. The high overall efficiency make these devices promising for applications such as remote sensing in the UV, particularly, when diode-pumped solid-state lasers will soon serve as pump.

1. Introduction

Many lidar applications require pulsed light sources emitting broadly tunable radiation in the ultraviolet spectral range with high peak and average power[1]. For example, by means of the DIAL (differential absorption lidar) technique using wavelengths in the range from 240-330nm various atmospheric species such as ozone, sulfur dioxide or toluene among others can be monitored.[2-4] Besides its wavelength and output energy essential features for the applicability of such a lidar transmitter are a high efficiency, reliability, simple handling and compactness. It is generally accepted that these design goals require all-solid-state laser sources.

There are only few solid-state laser materials that directly emit in the UV, and they require as pump the forth harmonic of a Nd:YAG laser with the associated disadvantages. Nevertheless, tunable UV radiation has been successfully generated using Ce:LiSAF or Ce:LiCAF lasers.[5,6]

Almost all other solid-state approaches use visible or near infrared radiation with frequency conversion such as second harmonic generation (SHG) or sum frequency mixing (SFM). In order to achieve a broad tuning range in the UV by means of these nonlinear optical techniques the light source should have a broad fundamental tuning range. In this respect, optical parametric oscillators (OPOs) pumped by the harmonics of Nd:YAG lasers offer substantial advantages over comparable tunable solid-state laser such as Ti:Sapphire or Cr:Alexandrite lasers. Another advantage of OPOs is the small timing jitter and a pulse width that is comparable to the pump. This is important for the UV generation by mixing the OPO radiation with the harmonics from the same pump source.

Potassium titanyl phosphate (KTP) and β-barium borate (BBO) are the OPO crystals that are presently best suited for the generation of pulsed radiation in the visible and near infrared spectral range. OPOs based on these nonlinear materials and pumped by the harmonics of Q-switched Nd:YAG lasers can nowadays be operated exceeding efficiencies of >30%.[7,8] Using Nd:YAG lasers as pump has the additional advantage of their well-known reliability and diode-pumping possibility.

In order to achieve UV wavelengths from OPOs, in most technical realizations the output of an OPO is frequency doubled by external second harmonic generation[9,10] with resulting overall optical efficiency of typically <1% only. The generation of UV radiation with wavelengths below ~330nm by means of SHG requires visible OPO radiation that can only be achieved with the third harmonic of the Nd:YAG laser as pump radiation of a BBO OPO. In contrast, using SFM of OPO radiation shorter wavelengths can be produced even starting from the second harmonic of the Nd:YAG laser as OPO pump resulting in a higher efficiency.

High pump fluences are necessary to achieve reasonable UV pulse energies because the conversion efficiency is a function of the product of the intensities of the incident waves. It could be demonstrated experimentally that the efficiency can be increased by intra-cavity doubling taking advantage of the high flux of the OPO signal inside the cavity.[11] In that work the signal wave of a KTP OPO was frequency-doubled giving tunable output from 380-520nm. This also holds true for SFM and has been demonstrated for cw-modelocked OPOs[12] as well as for a cw OPO[13] yielding wavelengths in the region of 589nm and 629nm, respectively.

The above listed advantageous properties of SFM make intra-cavity sum frequency mixing of nanosecond OPOs a very promising technique in order to efficiently generate continuously tunable ultraviolet radiation. It is therefore the goal of this work to investigate these OPOs in order to develop a versatile, efficient and compact UV light source for lidar applications.

2. Experimental

2.1 Intra-cavity sum frequency mixing of the OPO signal wave with the second harmonic of the pump

We chose the signal wave of a KTP OPO as tunable radiation and the second harmonic of the Nd:YAG laser as fixed frequency for the intra-cavity sum frequency mixing. KTP was chosen because its emission bandwidth is on the same order of magnitude than the acceptance bandwidth of the sum frequency mixing process in BBO.[14,15]

The set-up is depicted in Fig. 1a). As pump laser serves an injection-seeded flashlamp-pumped Nd:YAG laser delivering an energy of 410mJ at 1064nm. The repetition rate is 10Hz and the beam diameter is 6mm. The second harmonic output energy at 532nm is ~200mJ, and the pulse length is 6.5ns. The 532-nm pump beam is separated into an s- and p-polarized part by means of a half-wave plate and a polarizer. The s-polarized part is used to pump the type-II KTP OPO, whereas the p-polarized pulse is delayed for proper timing and serves as pump for type-I sum frequency mixing in a BBO crystal. The 75mm-long OPO cavity consists of three plane mirrors M1-M3 which are highly reflecting at the OPO signal wavelengths (700-830nm) and highly transmitting at 532nm. Mirror M3 additionally transmits the UV (300-360nm). A high-reflector for the 532nm-wavelength enables a double-pass pumping scheme for the OPO.

The advantage of using a variable polarizing pump beam splitter is that, for a given pump energy, the ratio of OPO pump to SFM pump can be optimized for maximum UV energy. With an optimized ratio and a pump energy of 205mJ a maximum UV energy of 19.5mJ could be achieved at a wavelength of 320nm. (See Fig. 2) This corresponds to a conversion efficiency of as much as 9.5% from the second harmonic to the UV or 4.8% from the fundamental of the Nd:YAG pump laser, respectively. Taking the UV transmission of the output coupler of ~90% into account the photon conversion efficiency is even higher. The device could be continuously tuned from 302-324nm with energies exceeding 10mJ (Fig. 2). This corresponds to an OPO signal wavelength range from 699-831nm. The tuning range in our set-up is limited by the mirrors and crystals used but should be extendable to 285-350nm.

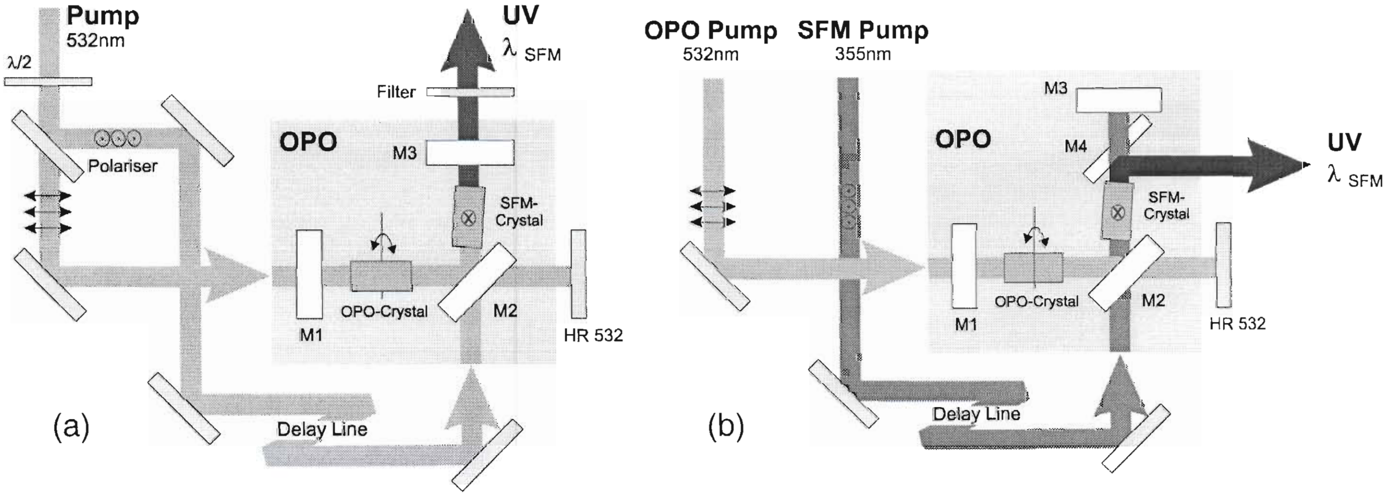

Figure 1:
(a) Experimental set-up of the OPO with intra-cavity sum frequency mixing when the 532nm second harmonic *of the Nd:YAG laser serves as pump wavelength for the SFM.*
(b) Experimental set-up of the OPO with intra-cavity sum frequency mixing when the 355nm third harmonic *of the Nd:YAG laser serves as pump wavelength for the SFM. An additional mirror M4 had to be implemented to reflect the UV beam off the cavity.*
For further details, see text.

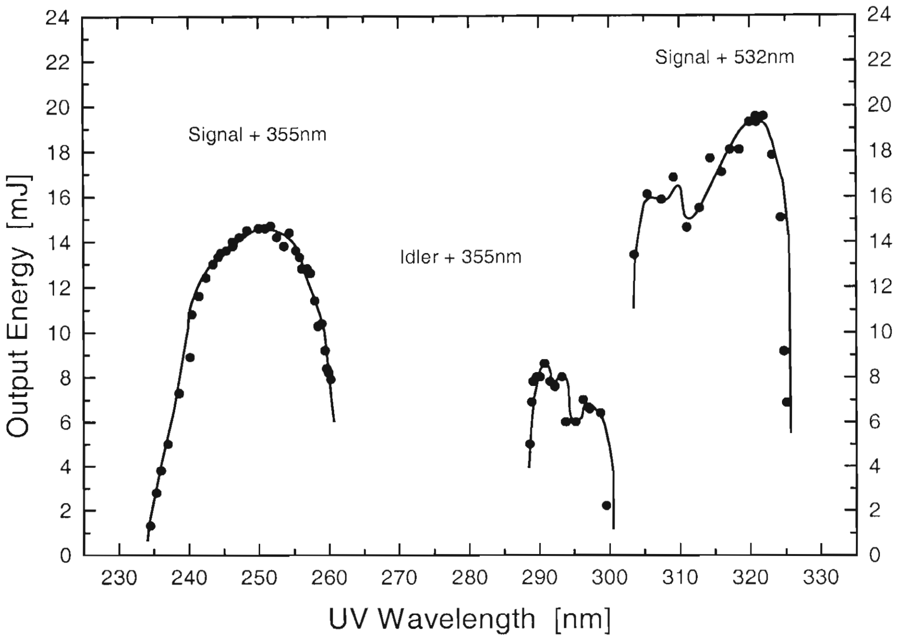

Figure 2:
Experimental tuning ranges of the investigated devices. The left curve was obtained by mixing the OPO signal wave (691-975nm) with 355nm. The tuning range was 234-260nm and the maximum UV output energy obtained was 14.7mJ.
The center curve was obtained by mixing the OPO idler wave (1545-1920nm) with 355nm. The tuning range was 288-300nm and the maximum UV output energy obtained was 8.6mJ.
The right curve was obtained by mixing the OPO signal wave (700-836nm) with 532nm. The tuning range was 302-325nm and the maximum UV output energy obtained was 19.5mJ.
For further details, see text.

The pulse width of the UV pulses was measured to be 5.2ns.[15] The bandwidth of the radiation could not be determined directly. But, according to the energy conservation conditions, the bandwidth of the sum frequency should be approximately 3 times less than the bandwidth of the second harmonic of the signal which was measured to be ~0.15nm. The energy stability of this OPO with intra-cavity SFM was very good. In a time series of 4min the rms noise of the energy was measured to be less than 1% (±3.2% p-p).[15]

2.2. Comparison to external frequency conversion

In order to demonstrate the advantages of the intra-cavity sum frequency mixing of a pulsed OPO this technique was compared to the conventional external sum frequency mixing. Therefore, the set-up depicted in Fig. 1a) was modified so that the external conversion could be performed under comparable conditions. The SFM crystal was removed from the cavity and, in order to efficiently extract the OPO signal from the cavity, high-reflector M3 was exchanged for an appropriate outcoupling mirror.

For the external sum frequency mixing the beams of OPO signal and 532nm SFM pump were overlapped by means of a dichroic beamsplitter and directed onto the BBO crystal for SFM. A UV energy of 9.7mJ at a wavelength of 315nm was achieved with a total (OPO pump plus SFM pump) energy of 197mJ.[15]

The OPO signal energy incident on the crystal was 22mJ and the SFM pump was 63mJ. Thus, the energy conversion efficiency is as high as 26%. In relation to the total second harmonic pump energy, the efficiency amounts to 4.9% and to 2.4% in relation to the fundamental energy, respectively. The comparison between the external (conventional) SFM and intra-cavity SFM clearly demonstrates the advantages of the intra-cavity mixing: its energy yield was twice as high under comparable conditions and so is the overall efficiency.

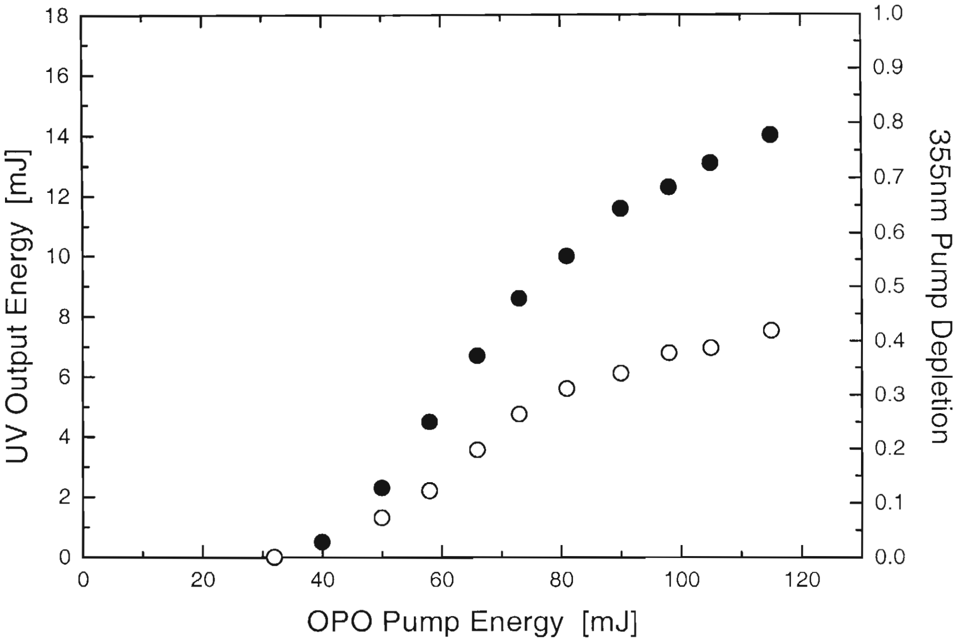

Figure 3:
UV output energy (full circles) and pump depletion of the 355 SFM pump radiation (open circles) as function of the OPO pump energy at a wavelength of 246nm. The maximum output energy was 14mJ. At the same time the depletion amounted to 42%. During this measurement the 355-nm energy was 33mJ per pulse.

2.3. Intra-cavity sum frequency mixing of the OPO signal wave with the third harmonic of the pump

With the described set-up, UV wavelengths below ~285nm cannot be generated. In order to extend the tuning range to shorter wavelengths the OPO signal wave has to be mixed with the third harmonic (355nm) of the Nd:YAG laser (Fig. 1b)). For these measurements the harmonic generator stage of the pump laser was modified so that the pump laser delivered ~120mJ of second harmonic and ~40mJ of third harmonic radiation at the same time. The horizontally polarized second harmonic was used to pump the KTP OPO crystal. A variable attenuator was implemented to vary the energy incident on the crystal. After an appropriate delay the vertically polarized third harmonic served as pump for the SFM. The OPO cavity is similar to the configuration depicted in Fig. 1a). Mirrors and crystal are the same. But as mirror M3 does not efficiently transmit wavelengths below 300nm an additional mirror (M4) had to be implemented. This mirror is highly transmittant for the OPO signal radiation but reflects wavelengths in the 250nm range off the cavity.

By means of this set-up, continuously tunable UV radiation was generated with wavelengths in the range of 234-260nm. This corresponds to an OPO signal wavelength range from 691-975nm. Figure 2 displays the wavelength dependence of the output energy of this device. During this measurement, the OPO pump energy was 100mJ and the SFM pump (355nm) amounted to 39mJ. The maximum achieved energy was 14.7mJ at 248nm.

Figure 3 shows the UV output energy at 246nm as a function of the OPO pump energy. A UV output energy of 14mJ was achieved with an OPO pump energy of 115mJ. At the same time the depletion of the 355-nm SFM-pump was higher than 40% (Fig. 3). During this measurement the 355nm SFM pump energy incident on the OPO cavity was 33mJ. The conversion efficiency corresponds to a value of 3.5% in relation to the fundamental or 7% in relation to the second harmonic wave, respectively. In a far field measurement the divergence of the sum frequency was determined to be less than 0.7mrad. This is mainly determined by the divergence of the pump laser which is ~0.5mrad.

Again, the obtained wavelength range is only limited by the mirror coatings used and the crystal angle range. The possible theoretical tuning range is as broad as 225-266nm using the signal radiation of the KTP OPO (614-1064nm). The generation of wavelengths >266nm can be performed by mixing the OPO idler radiation in an idler resonant cavity.

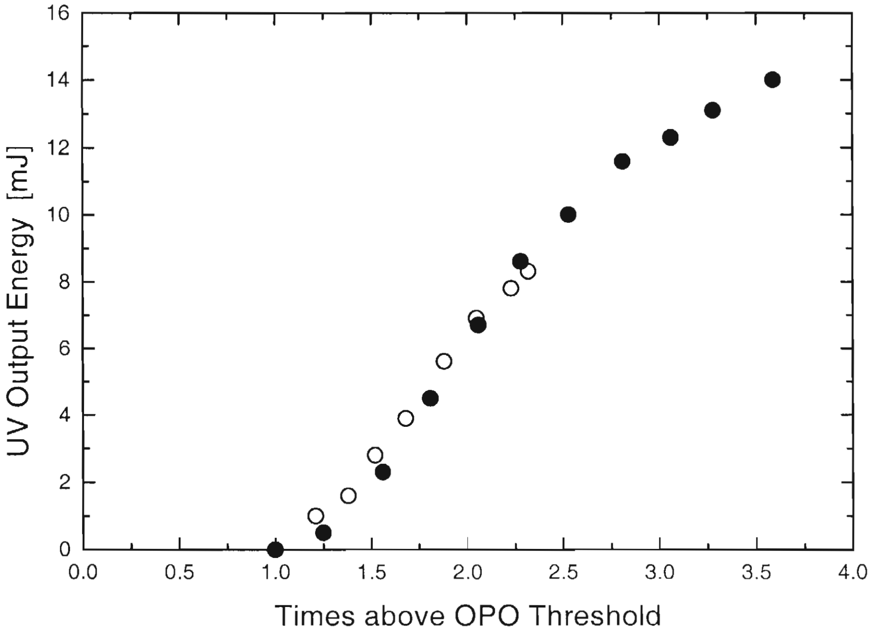

Figure 4:
UV output energy versus the number of times above OPO oscillation threshold for the intra-cavity sum frequency mixing of the 355-nm radiation with the signal wave (full circles) and the idler wave (open circles). The wavelength was 246nm and 291nm, respectively. The pulse energy at 355nm was 33mJ.

2.4. Intra-cavity sum frequency mixing of the OPO idler wave with the third harmonic of the pump

In order to demonstrate the sum frequency mixing of the OPO idler with the third harmonic of the pump the OPO cavity (Fig.1b) was equipped with mirrors that reflect in the wavelength range of 1550-1900nm in order to obtain an idler-resonant cavity. With this set-up the OPO could be tuned from 288-300nm (Fig. 2).

During this measurement, the OPO pump energy was ~125mJ and the SFM pump (355nm) amounted to ~33mJ. The maximum energy achieved was 8.6mJ at 291nm. This energy is considerably less compared to the latter measurement (signal wave +355nm). The reason is that the OPO crystal was not AR coated for the idler but for the signal wavelength range. Therefore the cavity losses and, consequently, the oscillation threshold of the OPO is much higher. However, when the UV energy is displayed as function of the pump energy expressed as the number of times above the OPO threshold the functional behavior is very similar to the latter case (see Fig. 4).

We can therefore expect that with optimized mirrors and crystal coatings we should be able to close the wavelength gap (Fig. 2) with comparable output energies.

3. Summary

In conclusion, we have devised nanosecond optical parametric oscillators with intra-cavity frequency mixing. These systems generate pulses in the UV spectral range with a maximum output energy of up to 20mJ. The maximum optical conversion efficiency from the Nd:YAG pump laser to the UV was as high as 4.7% corresponding to 9.5% in relation to the second harmonic of the Nd:YAG. A comparison with the external SFM revealed that intra-cavity SFM was the more efficient process. Under comparable conditions the output energies differed by a factor of 2. The high conversion efficiency is attributed to the high fluxes and to the well-defined phase fronts of the resonant signal radiation inside the OPO cavity.

At present, the tuning range of our OPOs is limited by the mirrors and crystals used to 234-260nm (signal wave + 355nm), 289-300nm (idler wave + 355nm) and to 300-326nm (signal wave + 532nm), respectively. Employing the whole KTP OPO signal tuning range will extend the UV tuning range to ~225-400nm.

These properties and in particular its high overall efficiency make this device promising for lidar applications in particular from mobile and airborne platforms. When diode-pumped solid-state lasers will replace the flashlamp-pumped systems as pump[11] the overall efficiency and average power of these OPO based tunable UV systems will further be increased.

Acknowledgements

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