Continuous-wave operation of a single-frequency optical parametric oscillator at 4–5 μm based on periodically poled $LiNbO_3$

M. M. J. W. van Herpen

Department of Molecular and Laser Physics, University of Nijmegen, 6525 ED Nijmegen, The Netherlands

S. E. Bisson

Combustion Research Facility, Sandia National Laboratories, Livermore, California 94550

F. J. M. Harren

Department of Molecular and Laser Physics, University of Nijmegen, 6525 ED Nijmegen, The Netherlands

Received June 18, 2003

We present a cw, Nd:YAG-pumped singly resonant single-frequency narrow-linewidth high-power optical parametric oscillator with idler tuning from 3.7 to 4.7 μm. In this spectral range the absorption of the idler wave in the $LiNbO_3$ crystal is significant, causing the oscillation threshold to increase with a subsequent decrease in output power from 1.2 W at 3.9 μm to 120 mW at 4.7 μm. The optical parametric oscillator's cavity was stabilized and mode-hop tuned with a rotatable solid etalon but with a subsequent reduction in idler power of as much as 50%. We demonstrated the usefulness for spectroscopy by recording the photoacoustic spectrum of a strong CO_2 absorption, using a 24-GHz continuous idler scan. © 2003 Optical Society of America

OCIS codes: 190.4360, 190.4410, 190.4970, 190.0190, 300.6340, 300.6390.

Optical parametric oscillators (OPOs) based on periodically poled lithium niobate (PPLN) are powerful tools for generating mid-infrared radiation because of their wide tunability, high power, high stability, and narrow linewidth.[1–8] Because the transparency of lithium niobate decreases at wavelengths above 4 μm, generation of radiation with longer wavelengths is difficult. However, there is strong interest in generation of long mid-infrared wavelengths for a number of applications such as spectroscopy and chemical sensing.

For pulsed OPO operation it has been shown that, because of the high parametric gain of the OPO, idler absorption loss in the PPLN crystal can be tolerated with operation extending far into the infrared absorption edge of $LiNbO_3$.[1,2] Recently, pulsed OPO operation was shown at idler wavelengths as long as 7.3 μm.[2] For longer wavelengths, pulsed difference-frequency generation can be used with other crystal types.[9,10]

In continuous-wave (cw) operation, OPO generation of idler wavelengths above 4 μm has continued to be a problem. Lowenthal[11] has made an analysis of the effect of idler loss on the cw OPO pump threshold and output power. His model was able to predict these values accurately for OPOs without idler absorption, but there has not yet been a validation of this model with strong idler absorption. In 1997 Myers and Bosenberg[3] described an OPO operating from 3.6 to 4.7 μm pumped by a multimode YAG source with a linewidth of ~2.2 GHz. They found that for wavelengths above 4 μm the idler output power dropped significantly, and they observed a "modest rise" in the oscillation threshold. Unfortunately, the oscillation threshold was not measured as a function of wavelength, and no information was given about pump depletion. In that study the authors did not stabilize the frequency of the OPO cavity with an intracavity etalon.

To be useful for many spectroscopic applications the OPO must have a narrow linewidth (<10 MHz), good stability, and broad tunability. Since the research of Myers and Bosenberg, to our knowledge no successful attempt has been made to develop a PPLN OPO operating with long idler wavelengths, probably because of lack of suitable pump sources. Here we present a Nd:YAG-pumped, cw, singly resonant OPO with single-frequency narrow-linewidth high-power output. The OPO can be continuously tuned over 24 GHz within its operating range of 3.7–4.7 μm. We demonstrated the usefulness for this system for spectroscopy by recording strong CO_2 absorption with photoacoustic spectroscopy.

The experimental setup is similar to a setup described previously.[7,8] The pump laser was a cw Nd:YAG source (Lightwave M6000) operating at 1064 nm, which generated as much as 11 W of single-mode output power. This laser had a small linewidth (5 kHz over 1 ms) with high frequency stability (50 MHz/h) and an excellent beam quality of $M^2 < 1.1$ in a TEM_{00} spatial mode. The frequency of the pump source could be continuously tuned over 24 GHz.[7]

The PPLN crystal used in this research had periods ranging from 25.9 to 28.7 μm and was initially operated at a temperature of 188.3 °C. The crystal was antireflection (R) coated for signal (<0.5% R at 1400–1500 nm), idler (<12% R at 3000–4850 nm), and pump (<0.5% R at 1064 nm) wavelengths. The OPO used a bowtie ring-cavity design with two flat mirrors and two curved mirrors, all coated (VLOC, New Port Richey, Fla.) for high reflectivity at the signal wavelength (≥99.8% R at 1350–1500 nm) and high transmission (T) for the pump (>90%% T at 1064 nm) and the idler (≥85% T at 3650–4850 nm) wavelengths. This configuration

0146-9592/03/242497-03$15.00/0

gave a singly resonant OPO, which was resonant for the signal frequency only. For spectroscopic applications, single-mode operation of this cavity could be enhanced by means of an intracavity 400-μm-thick uncoated YAG etalon (reflectivity, 8%). However, inserting this etalon into the OPO cavity caused a 50% reduction in idler power. The combination of a single-mode pump source and a single-mode ring cavity resonating at the signal wavelength produces a nonresonant but single-frequency idler beam with tuning characteristics that mirror those of the pump laser. Because the pump laser can be continuously tuned over 24 GHz, this tuning range can be used to tune the idler frequency as well. Additionally, the intracavity etalon can be rotated to mode-hop tune the OPO.[4]

The OPO oscillation threshold at 3.9 μm was found at 5 W, and with full pump power of 11 W an idler output of 1.2 W was achieved, with 70% pump depletion. The low pump depletion may be explained in part by low reflectivity of the cavity mirrors at the short signal wavelengths and by low transmission of the PPLN coatings at the edge of their specified range at 1.5 μm. In addition, it could also indicate that the cavity length was not optimized[8] or that there were thermal lensing effects.

The OPO was able to generate light with idler wavelengths ranging from 3.6 to 4.7 μm. For a pump power of 11 W and without the intracavity etalon, the output power varied from 1.2 W at 3.9 μm to 120 mW at 4.7 μm (Fig. 1), values that are similar to those of the output powers reported by Myers and Bosenberg.[3] Because idler photons with longer wavelengths have lower quantum energy, the output power is therefore correspondingly lower at longer wavelengths. However, the observed drop in output power with increased idler wavelength is due rather to intrinsic absorption of the idler wave in the PPLN crystal. At a pump power of 11 W and an estimated idler absorption of as much as 35%/cm,[11] thermal (lensing) effects can be quite large, causing a misalignment of the OPO cavity. Figure 1 shows the relationship of idler output power to the transparency of $LiNbO_3$. The oscillation threshold appeared to be stable near 5 W for idler wavelengths up to 4.2 μm, but for longer idler wavelengths the oscillation threshold increased significantly and became as high as 7.5 W at 4.7 μm (Fig. 2). At this wavelength the pump depletion is 10% with 11 W of pump power.

For the case of no idler absorption, Lowenthal's analysis[11] predicts an oscillation threshold of ~5 W at 4.5 μm, in agreement with our findings with low idler absorption. With an idler absorption of 35%/cm at 4.5 μm the oscillation threshold is predicted to increase to 7.3 W according Lowenthal's model, which is consistent with the observed value of 6.5 W. With 11 W of pump power Lowenthal predicts an idler output power of 1.0 W and a pump depletion of ~80% at 4.5 μm. However, our experimental values were much lower than this, with the idler output power near 110 mW (in agreement with Ref. 3) and a pump depletion of 10%. Because the pump depletion at 3.9 μm was also lower than predicted, we expected that is would also be lower at 4.5 μm. However, this was not enough to explain the large discrepancy from the model, especially since the model was able to accurately predict the oscillation threshold. The difference can be explained as being due to thermally induced focusing effects, which can significantly reduce OPO performance but are ignored in Lowenthal's analysis. With the high pump power of 11 W and the high idler absorption is it likely that these effects are present.

For a specific PPLN grating period and temperature, one could calculate the idler wavelength by using the Sellmeier equations of Jundt[12] (Fig. 3). To demonstrate the accuracy of this wavelength calculation in cases when there was significant absorption, we measured the absorption of a known CO_2 line in air (340 ± 10 parts in 10^6) at a wavelength of 4.201 μm (2380.72 cm^{-1}). For this measurement we set the coarse wavelength by translating the PPLN crystal to a period of 27.4 μm and operating the PPLN crystal at a temperature of 176 °C. Fine tuning to the peak of the CO_2 absorption line was achieved by mode-hop tuning[4] of the OPO with an intracavity etalon. Continuous pump tuning over 24 GHz was used to tune the idler frequency over the CO_2 absorption line. A maximum absorption of $52 \pm 4\%$ was found with a path length of 135 cm, giving an experimental absorption coefficient of $\alpha = 16 \pm 2\ atm^{-1}/cm^{-1}$. From the Hitran database a CO_2 absorption strength of $\alpha = 16.9\ atm^{-1}\ cm^{-1}$ was calculated at this wavelength.

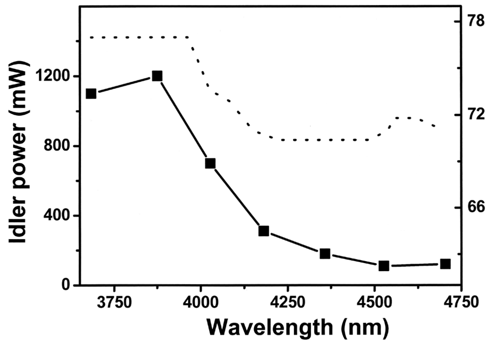

Fig. 1. OPO idler tuning with a multigrating crystal. The idler power (solid curve) correlates strongly with PPLN transparency (dotted curve).

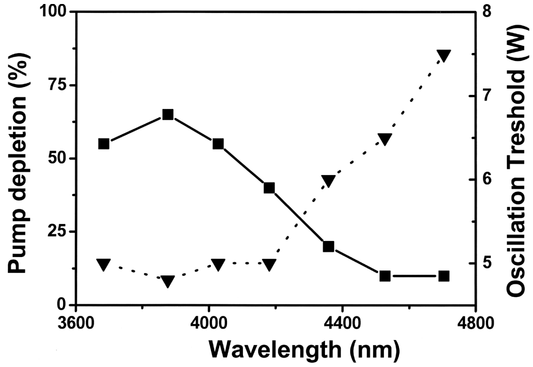

Fig. 2. Pump depletion (solid curve) and oscillation threshold (dotted curve) as a function of idler wavelength with 11 W of pump power.

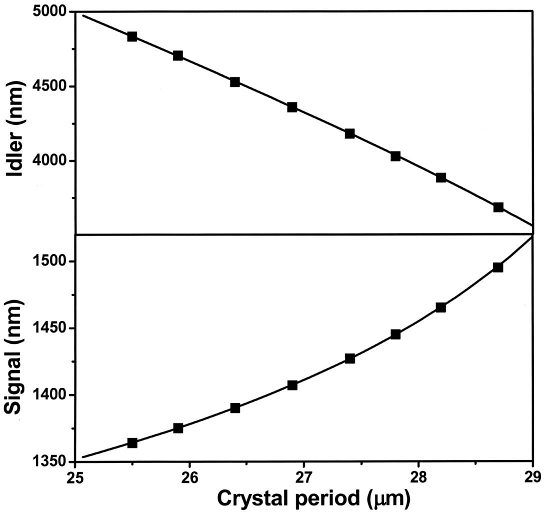

Fig. 3. Calculated signal and idler wavelengths at 188 °C. The squares indicate the crystal periods of the PPLN crystal. Changing the crystal temperature allows the full wavelength range from 3.8 to 4.8 μm to be covered.

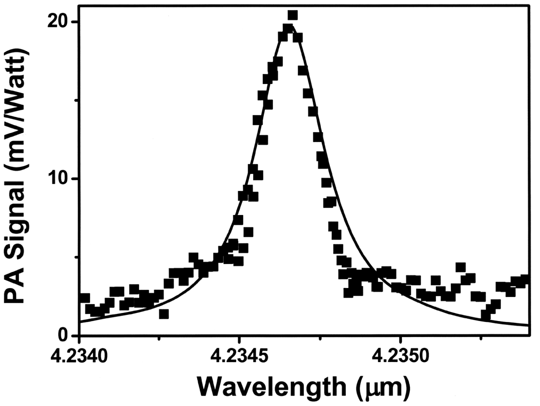

Fig. 4. Photoacoustic spectrum (squares) of the 4235-nm (2361.47 cm^{-1}) CO_2 line acquired by pump tuning the OPO over 24 GHz. For this measurement the CO_2 concentration was 7 parts in 10^6 in nitrogen. A Hitran calculation (solid curve) is shown for comparison.

As the calculated value is in agreement with the experimental value, this result confirms that the calculated wavelength can be used to set the OPO wavelength accurately.

The combined attributes of high power and broad tunability make this system an ideal source for spectroscopic and chemical sensing applications. To demonstrate the use of this system for spectroscopy in the 4.0–5.0-μm range we recorded photoacoustic spectra of a very strong CO_2 absorption (378.8 $\text{atm}^{-1}\ \text{cm}^{-1}$) at 2361.47 cm^{-1} (4235 nm).

To tune the OPO to this wavelength we used the same crystal period of 27.4 μm but with a temperature of 152.5 °C. Figure 4 shows a 24-GHz pump scan over the CO_2 absorption line at a concentration of 7 parts in 10^6 in nitrogen. The OPO cavity was covered with a box and flushed with pure nitrogen gas to minimize losses caused by atmospheric absorption. However, at the peak of the CO_2 absorption there was still a loss of at least 30% in the idler power that was due to residual absorption in the OPO environment. Despite these losses and the losses caused by the intracavity etalon, there was still 40 mW of idler power at the peak of the CO_2 absorption.

In conclusion, we have successfully developed a cw YAG-pumped singly resonant single-frequency high-power OPO operating from 3.7 to 4.7 μm. We found that, at longer idler wavelengths, absorption of the idler wave in the $LiNbO_3$ crystal significantly increased the oscillation threshold by as much as 2.5 W, in agreement with this part of Lowenthal's theoretical analysis.[11] For idler wavelengths longer than 3.9 μm the idler output power was considerably reduced as a result of intrinsic absorption within the $LiNbO_3$ crystal. Because of the high pump power and idler absorption, it was likely that there were induced thermal focusing effects. Our values of output power and pump depletion were not in agreement with Lowenthal's analysis, probably because that analysis excluded these effects. Despite this, the OPO cavity could be stabilized quite well and mode-hop tuned with a solid-state intracavity etalon. However, insertion of the solid etalon reduced the idler power by as much as 50%. For truly continuous tuning, the Nd:YAG pump laser was tuned over 24 GHz. These features make the system useful for spectroscopic applications, as we demonstrated by recording a strong CO_2 absorption with photoacoustic spectroscopy.

M. M. J. W. van Herpen's e-mail address is maartenh@sci.kun.nl.

References

1. P. Loza-Alvarez, C. T. A. Brown, D. T. Reid, W. Sibbett, and M. Missey, Opt. Lett. **24,** 1523 (1999).
2. M. A. Watson, M. V. O'Connor, P. S. Lloyd, D. P. Shepherd, D. C. Hanna, C. B. E. Gawith, L. Ming, P. G. R. Smith, and O. Balachninaite, Opt. Lett. **27,** 2106 (2002).
3. L. E. Myers and W. R. Bosenberg, IEEE J. Quantum Electron. **33,** 1663 (1997).
4. P. E. Powers, T. J. Kulp, and S. E. Bisson, Opt. Lett. **23,** 159 (1998).
5. M. E. Klein, C. K. Laue, D.-H. Lee, K.-J. Boller, and R. Wallenstein, Opt. Lett. **25,** 490 (2000).
6. W. R. Bosenberg, A. Drobshoff, and J. I. Alexander, Opt. Lett. **21,** 1336 (1996).
7. M. M. J. W. van Herpen, S. te Lintel Hekkert, S. E. Bisson, and F. J. M. Harren, Opt. Lett. **27,** 640 (2002).
8. M. M. J. W. van Herpen, S. Li, S. E. Bisson, S. te Lintel Hekkert, and F. J. M. Harren, Appl. Phys. B **75,** 329 (2002).
9. R. Haïder, A. Mustelier, Ph. Kupecek, E. Rosencher, R. Triboulet, Ph. Lemasson, and G. Mennerat, J. Appl. Phys. **91,** 2550 (2002).
10. O. Levi, T. J. Pinguet, T. Skauli, L. A. Eyres, K. R. Parameswaran, J. S. Harris, M. M. Fejer, T. J. Kulp, S. E. Bisson, B. Gerard, E. Lallier, and L. Becouarn, Opt. Lett. **27,** 2091 (2002).
11. D. D. Lowenthal, IEEE J. Quantum Electron. **34,** 1356 (1998).
12. D. H. Jundt, Opt. Lett. **22,** 1553 (1997).