2.2 Intra-cavity difference frequency generation in pulsed optical parametric oscillators

Hiroshi Komine***, Koji Suizu*, Takeshi Usami***, Koichiro Nakamura***, Kodo Kawase*, and Hiromasa Ito****

Abstract
Tunable 35- to 38-THz wave generation by difference frequency was demonstrated. An optical parametric oscillator (OPO) and intra-cavity difference frequency generator (iDFG) were used as the light source for difference frequency generation (DFG) of 30- to 38-THz waves. The iDFG consisted of two nonlinear periodically poled lithium niobate (PPLN) devices: one was for the OPO and the other was for DFG. The iDFG system could suppress the back conversion of pump light and had a higher conversion efficiency than conventional OPO systems. The iDFG system generated mid-infrared radiation at ~3.7 and 2.5 microns as a source of THz-wave generation using an AgGaSe2 crystal. A compact THz-wave source resulted, and changing the temperature of the PPLN permitted variation of the wavelength from 7.926 to 8.426 µm, which corresponds to 38-35 THz. This device was applied to the absorption spectroscopy of vapor gas.

1. Introduction
Nonlinear optical frequency conversion provides an effective way to generate coherent radiation in spectral regions that are not directly accessible by lasers. In particular, the advent of quasi-phase-matched (QPM) crystals, such as periodically-poled lithium niobate (PPLN) and new materials have advanced the techniques for generating tunable radiation from the near-infrared to the THz region [1]. Optical parametric interaction and frequency mixing in these media have demonstrated several paths to achieve useful power in the desired spectral band.

An optical parametric oscillator (OPO) pumped by a fixed-wavelength laser is a well-known method for generating tunable radiation at the signal and idler wavelengths. When pumped by a pulsed laser, an OPO produces signal and idler output pulses along with a significant amount of unconverted pump energy. However, recent advances in an OPO with intra-cavity difference frequency generation (DFG) have demonstrated that more efficient pump conversion is possible while increasing the idler output energy and generating a difference frequency radiation [2-4]. Such techniques yield simultaneous output pulses at two tunable mid-infrared wavelengths. DFG in an external frequency mixing crystal can then generate additional coherent radiation at longer infrared wavelengths. Thus, an OPO with an intra-cavity DFG (OPO-iDFG) offers several attractive capabilities for extending the spectral coverage using nonlinear frequency conversion.

---

* Photodynamics Research Center, RIKEN, 519-1399, Aramaki Aoba, Aoba-ku, Sendai 980-0845, Japan
** On Leave from TRW, One Space Park, Redondo Beach, CA 90278
*** Research Institute of Electrical Communication, Tohoku University, 2-1-1 Katahira, Aoba-ku, Sendai 980-8577, Japan
****
2. Background

The conversion efficiency in a pulsed OPO typically approaches a limiting value when pumped sufficiently above a certain energy required to reach oscillation threshold. Buildup time to reach the threshold accounts for a part of this behavior; however, once the oscillation starts, pump back conversion during the remainder of the pulse is responsible for the efficiency limit.

Back conversion occurs when the oscillating signal and idler waves traveling within the OPO crystal become sufficiently strong to cause complete pump depletion (CPD) locally in space and time. Parametric interaction continues beyond CPD to create new radiation at the pump frequency through sum frequency mixing of the signal and idler waves. This reversal of power flow leads to a complex temporal behavior, which ultimately prevents complete pump conversion over the pulse duration. Signal and idler pulses exhibit highly modulated temporal intensity spikes as a result of this dynamical process.

OPO-iDFG

DFG in an OPO cavity uses a second nonlinear crystal, as schematically shown in Figure 1. DFG creates a third wave at a frequency, $\omega_d$, given by the difference of the signal ($\omega_s$) and idler ($\omega_i$) frequencies (i.e., $\omega_d = \omega_s - \omega_i$). The parametric interaction of the signal and idler waves in the DFG crystal converts the signal power into the DFG and idler output, thereby creating two idler photons for each pump photon. This 2-for-1 process accounts for doubling the idler conversion efficiency, and it represents a key useful feature of OPO-iDFG.

![Fig. 1 Schematic diagram of OPO-iDFG device shown with a parametric gain section and an intra-cavity DFG section. Mirrors M1 and M2 form a singly resonant oscillator cavity for the signal wave. M1 transmits the pump radiation and reflects the signal wave. Pump radiation is unidirectional and drives the parametric gain section first. Signal and idler waves generated in the parametric gain section produce a difference frequency wave in the DFG section. M2 is an output coupler that transmits the idler and difference frequency radiation, but feeds back signal radiation.](image)

The signal power conversion in the DFG crystal also changes the dynamical behavior of the OPO itself due a nonlinear feedback mechanism. Both theoretical analysis [5] and experimental evidence [2-4] indicate that DFG can control the amount of signal power feedback in the OPO resonator and may significantly reduce pump back conversion. In such an OPO-iDFG device, signal wave oscillation starts as in a conventional OPO. When the signal and idler intensities grow substantially in the resonator, DFG begins to convert the signal power into additional idler power and difference frequency output. The loss of signal power creates a negative feedback that depends on the signal intensity.
When the relative strength of the parametric gain and difference frequency mixing falls within a certain range, the signal intensity in the resonator may evolve without driving the parametric interaction through the CPD crossing point. Under these conditions, pump depletion proceeds toward CPD but avoids back conversion. Remarkably, this dynamical feedback remains stable over a wide range of parametric gain. Consequently, the nonlinear feedback works even for a time-varying pump pulse from a Q-switched laser, and it can lead to efficient energy conversion into the idler and DFG output pulses while consuming signal energy in the process.

A third useful feature of the OPO-iDFG technique derives from the coherent mixing process in the DFG section. Since the idler wave is coherently amplified by the signal radiation in the DFG section, the 2-for-1 process retains the idler phase from the parametric gain section. Furthermore, the DFG pulse is temporally correlated with the idler pulse over its entire duration. This automatically assures idler and DFG pulses for further mixing in an external crystal.

PPLN OPO-iDFG Device:

PPLN crystals provide suitable media for QPM parametric gain and DFG mixing using polarization directions parallel to the crystal z-axis. Figure 2 shows calculated QPM period for the two processes as a function of signal wavelength for a Nd:YAG laser pump wavelength at 1064 nm.

![Graph: PPLN OPO-iDFG QPM period as a function of signal wavelength](image)

**Fig. 2** PPLN OPO-iDFG QPM period as a function of signal wavelength for 1064 nm pump. Also plotted are idler and DFG wavelength.

Signal wavelength tuning from about 1.5 to 1.59 microns yields idler wavelength from about 3.7 to 3.2 microns, respectively. The corresponding DFG wavelength ranges from 2.5 to 3.2 microns. Signal wave tuning from about 1.6 to 1.7 microns yields idler wave in the 3.2
to 2.7 micron range with DFG wavelength of 3.2 to 4.5 microns. Further mixing of these idler and DFG waves in a suitable external crystal can generate coherent pulsed radiation from 8 microns to THz waves.

3. Experimental results

Pulsed intra-cavity OPO-DFG (OPO-iDFG) using two periodically poled lithium niobate (PPLN) crystals and cascaded mid-infrared generation was demonstrated using AgGaSe2. The OPO-PPLN crystal operated in the usual manner by converting pump photons ($\omega_p$) into signal ($\omega_s$) and idler ($\omega_i$) photons ($\omega_p = \omega_s + \omega_i$). The DFG-PPLN crystal was designed to phase match the signal and idler waves, to produce a difference frequency photon ($\omega_d = \omega_s - \omega_i = 2\omega_s - \omega_p$) and an additional idler photon. Since the signal photon is generated in the OPO, but is consumed in the nonlinear DFG process, back conversion is minimized and pump depletion is improved. The output of the OPO-iDFG ($\omega_i$ and $\omega_d$) is used to generate inter-cavity DFG radiation ($\omega_D = \omega_d - \omega_i$) using an AgGaSe2 crystal. By changing the temperature of the PPLN crystals, the wavelength of the OPO-iDFG radiation ($\omega_i$ and $\omega_d$) changes, and consequently changes the wavelength of the inter-cavity DFG radiation ($\omega_D$). The experimental setup and energy diagrams of each process are shown in Fig. 3.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig3}
\caption{The experimental setup and energy diagrams for each process involved in PPLN-OPO-iDFG and inter-cavity DFG using AgGaSe2.}
\end{figure}

For the OPO/DFG process, we used 0.5-mm-thick PPLNs. The pump source was a Q-switched Nd:YAG laser, operating at a 50-Hz repetition rate. The laser produced a smooth pulse envelope with a FWHM of 25 ns. We chose a singly resonant oscillator cavity for the OPO/DFG.

Operating the OPO-iDFG at a pump energy of 1.4 mJ, the idler ($\omega_i$) and DFG ($\omega_d$) energies were 0.2 mJ at a 50-Hz repetition rate. By changing the crystal temperature from 130-150°C, the idler and DFG wavelengths were tuned from 3.705 to 3.670 $\mu$m and from 2.501 to 2.533 $\mu$m, respectively. These wavelengths correspond to DFG wavelength from 7.693 to 8.179 $\mu$m ($\omega_D = \omega_d - \omega_i$) using an AgGaSe2 crystal.

As seen in Fig. 4, with angle tuning of the AgGaSe2 crystal, the experiment and calculated values were in very good agreement. By changing the temperature of the PPLN crystals from 140-160°C, $\omega_D$ was tuned from 7.926 to 8.426 $\mu$m, which correspond to 38-35 THz.
Fig. 4  Angle-tuning of an AgGaSe$_2$ crystal and the calculated curve. The pump energy was 1.2 mJ and the temperature of the PPLN crystals was 145 °C.

Figure 5 shows an example of the absorption spectrum measurement of vapor. The gas cell used was a 50-cm-long brass light pipe with CaF$_2$ windows at both ends. The intensities of the DFG radiation that penetrated the gas cell in air at atmospheric pressure (contained vapor) and under vacuum were measured, at different PPLN crystals temperature. The observed absorption line agreed with the reference value well [6].

Fig. 5  The absorption spectrum of vapor. The temperature of the PPLN crystal was changed from 145-150°C. The pump energy was 1.2 mJ.
4. Summary

Tunable 35- to 38- THz-wave generation by difference frequency generation of OPO-iDFG output radiations was demonstrated. The OPO-iDFG system generated mid-infrared radiations at ~3.7 and 2.5 microns as a source of THz-wave generation (using an AgGaSe₂ crystal), and suppressed the back conversion of pump light. A compact THz-wave source resulted, and changing the temperature of the PPLN permitted variation of the wavelength.

5. Acknowledgments

The authors thank C. Takyu for his excellent work coating the crystal surfaces, and T. Shoji for polishing the crystals superbly.

References