Continuous-wave frequency tripling and quadrupling by simultaneous three-wave mixings in periodically poled crystals: application to a two-step 1.19–10.71-μm frequency bridge

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Received April 3, 1997

We observed cw third-harmonic generation in a periodically poled $LiNbO_3$ crystal by cascading optimally phase-matched second-harmonic and sum-frequency generation. Other processes, such as fourth-harmonic generation, are allowed by the flexibility of quasi-phase matching. We demonstrate a divide-by-nine (1.19–10.71-μm) frequency chain that uses only two lasers.

Precise measurement of optical frequencies requires connecting laser frequencies to well-known standards by means of a frequency chain. Because of the large frequency intervals involved, these systems can be extremely complex, but they are required for future optical clocks and tests of fundamental physics.[1] Therefore there is a need for new and efficient optical mixing schemes compatible with cw, low-power frequency-stabilized lasers. Our interest is also in frequency connections in a 3:1 ratio, which would bridge gaps between excellent existing standards such as the CO_2 laser locked on OsO_4 at 10 μm, the He–Ne laser locked on CH_4 at 3.39 μm, and the Nd:YAG laser locked on I_2 at 1 μm.

Nonlinear mixing in crystals is a good way to connect optical frequencies.[2] One promising technique is quasi-phase matching[3,4] (QPM) in periodically poled (PP) crystals,[5] such as $LiNbO_3$ (LN), $KTiOPO_4$ (KTP), and $RbTiOAsO_4$ (RTA). QPM has well-known advantages such as access to the largest nonlinear coefficients, suppressed walk-off, and great flexibility in the choice of the wavelengths involved in the mixing.

Because of these properties, simultaneous QPM of different interactions is more probable than one might at first suppose. We envisaged third-harmonic generation (THG) by cascading[6] second-harmonic generation (SHG; $\omega \mapsto 2\omega$) and sum-frequency generation (SFG; $\omega + 2\omega \mapsto 3\omega$), which of course realizes a 3:1 frequency connection. Another possibility (using a single input beam as well) is fourth-harmonic generation (FHG) by cascading SHG twice. Considering simple poling (50% duty-cycle period) and first-order collinear QPM[4] as well as birefringent phase matching (BPM),[7] and allowing different polarizations, we find 12 THG and 10 FHG coincidences in PP LN (Fig. 1 and Table 1), 8 and 2 coincidences in PP KTP, and 14 and 5 in PP RTA (Table 2).

Given our low initial power $P(\omega)$, the THG power is $P(3\omega) = \eta_{SHG}\eta_{SFG}P(\omega)^3$, where $\eta_{SHG,SFG}$ are the SHG and SFG efficiencies, each of the form $[\chi^{(2)}\mathrm{sinc}(\Delta\mathbf{k}\cdot\mathbf{l}/2)]^2$. $\Delta\mathbf{k}$ is the phase-mismatch vector and $\mathbf{l}$ is a vector of modulus the effective crystal's length, perpendicular to the walls of the poled domains. Although this model takes into account noncollinear QPM (angle tuning)[4] and the temperature dependence of the

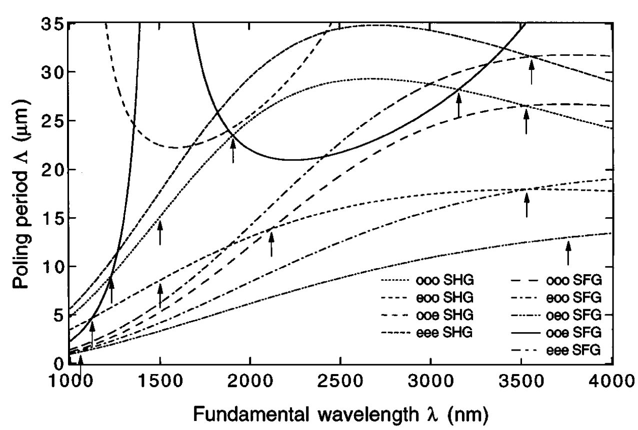

Fig. 1. QPM curves for THG in PP LN: The optimal poling period Λ is calculated versus the input wavelength λ for SHG and SFG and several polarizations. These curves are based on the Sellmeier coefficients[8] that gave predictions closest to our experimental results at room temperature. Arrows point to the THG coincidences. As in the tables, polarizations (o, e) are given for $\omega\omega 2\omega$ and $\omega 2\omega 3\omega$.

Table 1. THG and FHG Coincidences in PP LN[a]

THG in PP LN				FHG in PP LN			
λ_{in}	λ_{out}	Λ	Polarizations	λ_{in}	λ_{out}	Λ	Polarizations
1056	352	∞/1.2	*ooe/oeo*	1755	439	11.0	*eoo/ooe*
1122	374	4.6	*eoo/ooe*	1897	474	23.3	*ooo/ooe*
1231	410	9.0	*ooo/ooe*	2113	528	13.8/∞	*eoo/ooe*
1500	500	8.5/∞	*eoo/ooe*			26.4/∞	*ooo/ooe*
		5.3/∞	*ooo/ooe*	2524	631	29.1	*ooo/ooe*
1907	636	23.5	*ooo/ooe*	3240	810	17.8	*eoo/ooo*
2118	706	13.9	*oeo/ooo*	3759	940	∞/27.1	*ooe/eee*
3158	1053	28.2	*ooo/ooe*	3893	973	24.8	*ooo/ooe*
3527	1176	18.0	*eoo/eoo*	3941	985	24.5	*ooo/ooo*
		26.6	*ooo/ooo*	3979	995	29.2	*eee/eee*
3561	1187	31.6	*eee/eee*				
3759	1253	∞/13.0	*ooe/oeo*				

[a] $\lambda_{in,out}$ in nanometers; poling period Λ (or $\Lambda_{SHG}/\Lambda_{SFG}$) in micrometers. $\Lambda \to \infty$ indicates BPM. Polarizations (o, e) are given for $\omega\omega 2\omega/\omega 2\omega 3\omega$ for THG and for $\omega\omega 2\omega/2\omega 2\omega 4\omega$ for FHG. Nonlinear efficiencies $\propto (d_{22})^2\,(ooo) < (d_{31})^2\,(ooe, oeo, eoo) \ll (d_{33})^2\,(eee)$. The boldface number is the wavelength at which the experiment was performed (see text).

Table 2. THG and FHG Coincidences in PP KTP and PP RTA[a]

λ_{in}	λ_{out}	Λ	Polarizations
THG in PP RTA			
1143	381	∞/1.7	*zxx/xxz*
		∞/5.9	*zxx/zxx*
1244	415	∞/2.2	*zyy/yyz*
1368	456	6.0/∞	*xxz/xzx*
1492	497	7.4/∞	*yyz/yzy*
1720	573	54.7	*zxx/zxx*
1916	639	75.1	*zyy/zyy*
2196	732	67.2/∞	*zxx/zxx*
3312	1104	1290	*zyy/zyy*
3394	1131	∞/12.4	*zyy/yyz*
3603	1201	∞/∞	*zxx/zxx*
3604	1201	∞/12.3	*zxx/xxz*
3630	1210	455	*zyy/zyy*
3658	1219	43.4	*zzz/zzz*
FHG in PP RTA			
2424	606	12.6	*xxz/zzz*
2501	625	13.9	*yyz/zzz*
3394	849	∞/8.9	*zyy/yyz*
3604	901	∞/17.3	*zxx/xxz*
4091	1023	40.1	*zzz/zzz*
THG in PP KTP			
1082	361	∞/4.6	*zyy/zyy*
1213	404	4.6/∞	*xxz/xzx*
1325	442	5.6/∞	*yyz/yzy*
1473	491	36.5	*zxx/zxx*
1623	541	47.8	*zyy/zyy*
1824	608	40.9/∞	*zxx/zxx*
2052	684	∞/6.0	*zyy/yyz*
		59.0/∞	*zyy/zyy*
FHG in PP KTP			
2161	540	9.8	*xxz/zzz*
2227	557	10.8	*yyz/zzz*

[a]Same as in Table 1. Nonlinear efficiencies $\propto (d_{31})^2(xxz, xzx, zxx) < (d_{32})^2(yyz, yzy, zyy) \ll (d_{33})^2(zzz)$. Note, in RTA, the THG and FHG of the He–Ne/CH_4 standard (3392 nm) and the rare BPM–BPM THG (3603 nm).

indices[8,9] and of the length[10] of the crystal, it did not fit our experimental results well, probably because the Sellmeier coefficients[8,9] are not accurate beyond 3 μm. Still, one set of coefficients[9] enabled us to predict qualitatively that higher temperatures should give THG at longer wavelengths and incidences closer to normal, a prediction confirmed by the experiment.

We demonstrated experimentally THG coincidence *eee/eee* in PP LN (Fig. 1, Table 1). A CO overtone laser,[11,12] emitting on its $v' = 30 \to v = 28$ band (3.54–3.61 μm) with an output power of $\leq$250 mW, was focused to a 50-μm waist in our 20 mm $\times$ 15 mm $\times$ 0.5-mm PP LN sample. The crystal was poled with a period $\Lambda = 31.5\ \mu$m and antireflection coated at 1.8 and 1.2 μm. The output SHG at 1.8 μm was measured with an InGaAs photodiode with a 2.2-μm cutoff. The efficiency inside the crystal was $P(2\omega)/P(\omega)^2 \geq 5.4\ 10^{-4}\ \mathrm{W}^{-1}$. The third-harmonic signal was then detected and measured with an optical spectrum analyzer. The light was brought to it by a 400-μm-diameter multimode fiber. This large fiber core was most convenient for optimizing the mixing (crystal angles, focusing) for maximum THG signal.

By measuring the THG efficiency for different CO laser lines, we determined that the optimum THG coincidence point occurs at a shorter wavelength than expected from the theory (Table 1). At room temperature the THG coincidence point for our crystal is at <3.54 μm, which is in the gap between the 29 → 27 and the 30 → 28 bands of our CO overtone laser. Even so, ~200 mW out of the CO laser still produces a maximum THG power of ~0.5 nW at 22 °C and ~15-deg incidence. We were then able to temperature tune the optimum coincidence point to 3.56 μm [$P_{28}(9)$], for a temperature of 133 °C and nearly normal incidence. We obtained a THG power of ~7.3 nW (sufficient for phase-locking purposes) for a CO power of 195 mW (Fig. 2), which corresponds to a THG efficiency inside the crystal of $P(3\omega)/P(\omega)^3 \leq 10^{-6}\ \mathrm{W}^{-2}$. This optimized efficiency corresponds to normal incidence, i.e., maximum interaction length (collinear QPM) in our crystal.

As the first stage in using THG for optical frequency synthesis we made a two-step, divide-by-nine frequency conversion between 10.71 and 1.19 μm. The CO overtone laser, emitting at 84 THz on $P_{28}(11)$ (3.57 μm),

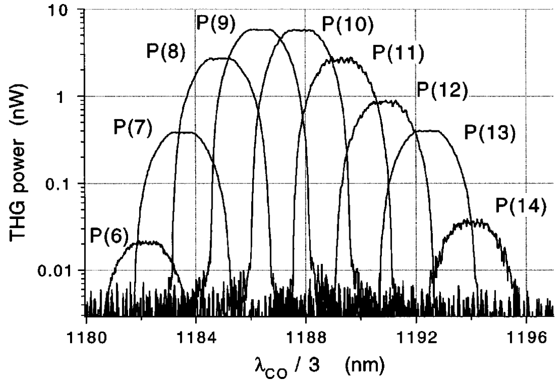

Fig. 2. THG power, at 133 °C, for several CO overtone frequencies. The width of the signals is the resolution of the optical analyzer, fixed by the fiber's diameter (input slit).

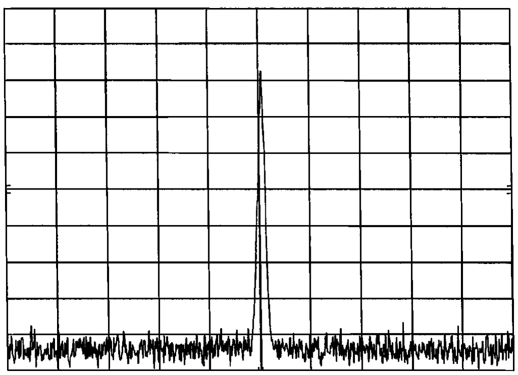

Fig. 3. MIM-diode voltage of beat note $f_{CO} - 3f_{CO_2}$. Vertical linear scale. Center frequency, 796 MHz; scan, 10 MHz; resolution and video bandwidths, 100 and 30 kHz, respectively.

was frequency tripled in PP LN to 252 THz (1.19 μm). It was also connected to a $^{12}C^{18}O_2$ laser emitting on $P_I(40)$ (28 THz, 10.71 μm) by use of a metal-insulator-metal (MIM) diode; the electrical IF signal from the diode corresponded to the beat note between the tripled CO_2 laser and the CO laser.[12] The two laser beams were focused on the MIM diode at orthogonal angles, as has proved to be optimum.[13] The laser powers and the resultant rectified voltages in the MIM were 70 mW and 0.5 mV for the CO laser and 200 mW and 5 mV for the CO_2 laser. We obtained a 796-MHz beat note with a signal-to-noise ratio of $\sim$25 dB in a 100-kHz detection bandwidth (Fig. 3), which should be adequate for phase locking the CO laser. CO_2 lasers have now been stabilized to the hertz level on the short term, with subhertz Flicker plateaus and reproducibilities of < 10 Hz.[14] Phase locking a CO laser to such a reference would yield a short-term stability of $\sim$10 Hz at 1.19 μm.

We have demonstrated a new use of QPM in PP crystals for optical frequency mixing. One can remove the constraint of finding coincidences by juxtaposing different gratings in the same crystal, each grating corresponding to each desired mixing. This procedure can also lead to an optical equivalent of rf diode mixers that generate the sum, difference, and second harmonics of two input frequencies. We have observed all four of these mixings (occurring at different angles and QPM orders) out of a single-grating PP LN crystal with inputs from an 800-nm diode and a 1064-nm Nd:YAG laser. More generally, Fejer *et al.* proposed an elegant design method of phase matching curves by Fourier analysis of spatial poling frequencies.[4] Hence QPM opens many new possibilities in optical frequency synthesis and high-resolution spectroscopy, especially because these applications do not usually require high power. Optical beat notes can be detected with $\geq$1 nW of power, and ultrahigh-sensitivity saturation spectroscopy can now be achieved with low initial power by use of buildup in high-finesse optical resonators.[15]

We thank L. E. Myers, R. W. Fox, S. Waltman, A. Bredsford, K. Petrov, and J. L. Hall for significant contributions to this research, which is supported in part by the U.S. Air Force Office of Scientific Research.

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