Pulsed injection-seeded optical parametric oscillator with low frequency chirp for high-resolution spectroscopy

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An injection-seeded optical parametric oscillator (OPO), based on periodically poled KTiOPO$_4$ and pumped by a frequency-doubled, nanosecond-pulsed Nd:YAG laser, generates continuously tunable, single-longitudinal-mode, pulsed output at ~842 nm for high-resolution spectroscopy. Optical-heterodyne measurements show that the OPO frequency chirp increases linearly with detuning from the free-running (unseeded) OPO frequency and can be maintained as low as 10 MHz. Other factors affecting chirp are identified.   © 2003 Optical Society of America

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High-performance pulsed (rather than cw) sources of intense coherent radiation are needed in many high-resolution spectroscopic applications, for example, those using pulsed tunable lasers to generate vacuum-ultraviolet radiation by nonlinear-optical frequency conversion.[1] In this Letter we report an injection-seeded optical parametric oscillator (OPO) as a well-characterized tunable narrowband source of nanosecond-pulsed coherent radiation for such applications.[2]

Ideally, pulsed spectroscopic measurements require a source of coherent light with continuous tunability, good pulse-to-pulse stability, well-defined optical phase, and optical bandwidth approaching the Fourier-transform limit.[3] It is important in this context to be able to control and minimize instantaneous frequency perturbations that often arise in intense, pulsed coherent light sources. An optical-heterodyne (OH) technique[4] can be used to measure the optical phase properties of pulsed, tunable coherent light, as has been done in characterizing high-resolution spectroscopic systems comprising a single-longitudinal-mode (SLM) cw laser that is pulse amplified by dye-laser media.[1,3,5]

Here we adapt this OH approach to study a solid-state nonlinear-optical system, namely, a nanosecond-pulsed SLM-tunable OPO based on periodically poled KTiOPO$_4$ (PPKTP) and injection-seeded by a cw tunable diode laser (TDL). This approach enables us to examine, on a nanosecond time scale, how perturbations in the frequency of the OPO output, both within a single pulse and from pulse to pulse, depend on experimental parameters such as phase mismatch Δk of the OPO process, offset of the OPO cavity resonance frequency from the injection-seeding frequency, and pump-laser fluence. In nonlinear-optical media, frequency perturbations (often referred to as chirp when the changes in instantaneous frequency during a pulse are linear or monotonic) can arise from phase modulation through the nonlinear refractive index,[3,6] from cascaded $\chi^{(2)}$ interactions,[7] from chirp in the pump-laser pulse itself, and from cavity-pulling effects when the OPO cavity resonance frequency is displaced from the injection-seeding frequency.

The apparatus is shown in Fig. 1. The PPKTP OPO is pumped at 532 nm by a frequency-doubled Nd:YAG laser (Spectra Physics GCR-250) that delivers SLM Q-switched pulses of duration ~8 ns, full width at half-maximum (FWHM). The OPO ring cavity is resonant at the signal wavelength (~842 nm in this work), with all OPO resonator mirrors anti-reflection coated for 532 nm; in the wavelength range 800–900 nm, mirror M1 is ~80% reflective and mirrors M2–M4 are high (>99%) reflectors. The PPKTP crystal (Raicol Crystals; 1 mm × 2 mm × 20 mm; 9.35-μm grating period) is mounted in a compact oven that can be heated to 250 °C. The unseeded OPO signal wavelength can be tuned from 815 to 877 nm by variation of the PPKTP oven temperature
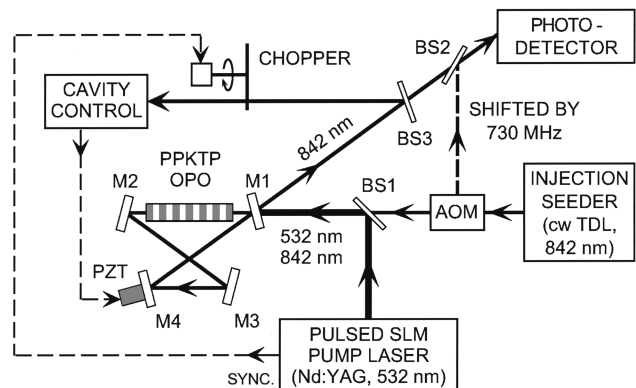

Fig. 1. Schematic of the TDL-seeded ring-cavity PPKTP OPO and the optical-heterodyne measurement system. M1–M4, mirrors; BS1–BS3, beam splitters; AOM, acousto-optic modulator; PZT, piezoelectric translator; SYNC., sychronization; other abbreviations defined in text.

from 200 to 20 °C. The OPO signal output beam has a near-Gaussian spatial beam profile. Typically, the free-running (unseeded) PPKTP OPO generates ~ 20 μJ/pulse of signal output with a pump-laser pulse energy of ~ 0.1 mJ (approximately three times the OPO threshold, 35 μJ/pulse).

Our previous work[2,8,9] showed that injection seeding by a cw SLM TDL can generate OPO output that is also SLM and continuously tunable with an effective optical bandwidth of ~ 100 MHz. The TDL used here (New Focus 6316) has a tuning range 834–851 nm, which defines the wavelength range over which the SLM OPO can be operated. Our intensity-dip locking scheme[8,9] is implemented via beam splitter BS3, a chopper, a piezoelectric translator, and cavity-control electronics. The vacuum wavelengths of the injection seeder and the OPO signal output are monitored by a wavemeter (Burleigh 4500-1).

To make OH beat measurements, we deflect a portion of the TDL injection-seeder output and frequency shift it by +730 MHz in an acousto-optic modulator (Brimrose TEM-100-30). The unshifted zero-order beam transmitted by the acousto-optic modulator is used to seed the OPO and is combined collinearly with the pump beam by beam splitter BS1. The deflected, frequency-shifted cw light is combined collinearly with the pulsed 842-nm OPO signal radiation by BS2 and focused onto a 1-GHz photodiode (New Focus 1601) to produce a heterodyne beat signal that is recorded by a 2-GHz digital oscilloscope (Tektronix TDS794D).

To extract optical phase information, we first Fourier transform the recorded OH beat waveform into the frequency domain. The positive-frequency portion of the resulting complex-valued Fourier spectrum comprises two bands, one of which (at lower frequency) is associated with the intensity envelope of the OPO output pulse. The other band (around the acousto-optic modulator frequency) is associated with the OH beat waveform, and one can select it by use of a bandpass filter (e.g., Blackman-type) while suppressing all other frequency components outside the filter's bandwidth. The filtered spectrum is then backtransformed to yield the temporal evolution of the amplitude and phase of the original pulsed OPO radiation field.[3] The time derivative of the phase yields the instantaneous frequency f_{inst} of the OPO pulse. Before the filtered spectrum is backtransformed, its band center is shifted toward zero, away from the beat frequency (~ 730 MHz), thereby displacing the mean value of the time derivative of phase to avoid associated numerical artifacts.

Figure 2 depicts a typical OPO signal pulse, the OH beat waveform, and the instantaneous-frequency profile (f_{inst} plotted against time) extracted with the Fourier-transform algorithm. From the OPO signal pulse profile, we derive a single-shot Fourier-transform bandwidth of 60 $\pm$ 5 MHz FWHM, consistent with that for an ideal 7.5-ns FWHM Gaussian pulse. The chirp can be defined by a linear fit to the f_{inst} profile during the time interval between the 10%-intensity levels of the OPO signal pulse. Alternatively, as in Fig. 2, we can define the chirp as the difference Δf_{inst} between maximum excursions of

f_{inst} over the same time interval; Δf_{inst} is negative if the positive excursion precedes the negative excursion. Both definitions yield similar results, but the latter is advantageous when large excursions in f_{inst} occur with the slope of the linear fit close to zero.

Figure 3 shows how the linear fit to the f_{inst} profile and Δf_{inst} vary as a function of the seeded OPO signal output wavelength. To record these data, we set the PPKTP crystal temperature at 125 °C and the pump-pulse energy was 45 μJ (1.3 times threshold for the unseeded OPO). The vertical solid line depicts the center wavelength, λ_{free}, of the signal output from the free-running OPO (841.56 nm). The magnitude of Δf_{inst} increases as the TDL-seeded signal wavelength of the OPO moves away from λ_{free}. The sign of Δf_{inst} is such that, during the evolution of the OPO pulse, f_{inst} shifts toward the OPO's free-running center frequency. This is consistent with the theory of Planken et al.[6] for a picosecond-pulsed optical parametric amplifier (OPA), in which the sign of the chirp [attributable to $\chi^{(2)}$ effects] is determined by the sign of the nonlinear-optical phase mismatch, Δk.
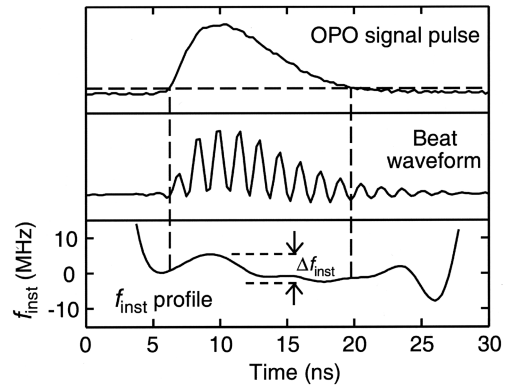

Fig. 2. Top, OPO signal pulse with 10%-intensity points marked by dashed lines. Middle, OH beats between frequency-shifted TDL seed light and the OPO pulse. Bottom, instantaneous-frequency profile. The chirp can be estimated from either a linear fit or, as shown here, from the maximum frequency excursions Δf_{inst} in the 10%-intensity range.
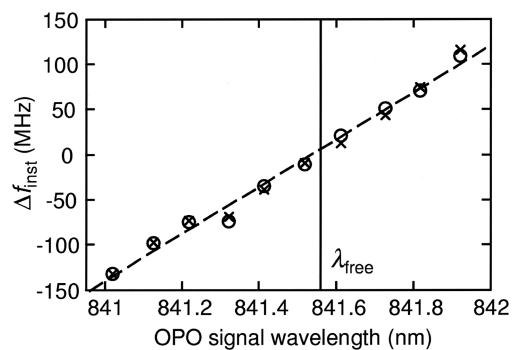

Fig. 3. Frequency chirp as a function of injection-seeded OPO signal wavelength. Circles, Δf_{inst} chirp values; crosses, chirp from a linear fit to f_{inst} within the 10% intensity range; dashed line, least-squares fit to the circles, with a slope of 261 $\pm$ 9 MHz/nm; vertical line, free-running OPO center wavelength λ_{free} (841.56 nm).
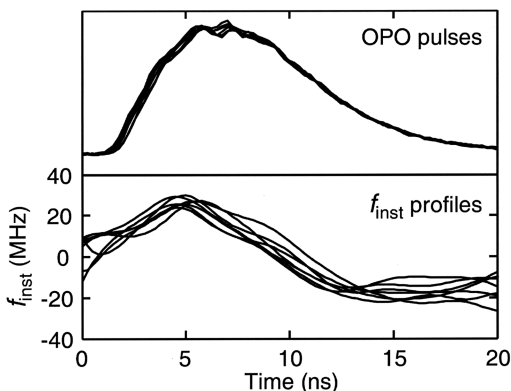
Fig. 4. Seven individual temporal OPO pulse profiles with instantaneous-frequency profiles.

A straight-line least-squares fit to the Δf_{inst} data (circles) in Fig. 3 has a slope of 261 ± 9 MHz/nm, corresponding to a chirp of 0.62 MHz toward λ_{free} for every 1 GHz that the seeded OPO is detuned from λ_{free}. The minimum value of Δf_{inst} occurs when the seeded OPO signal wavelength coincides with λ_{free} (within the ± 0.02-nm measurement variation).

In the case of dye-laser amplifiers, one needs to alter the composition of the dye solution to adjust the minimum chirp wavelength.[5] Here, by contrast, one can adjust the value of λ_{free} to minimize the chirp simply by varying the PPKTP crystal temperature. For one of the OH measurements at 125 °C shown in Fig. 2, the TDL-seeded OPO signal wavelength differs by only -0.04 nm from the corresponding value of λ_{free}, and Δf_{inst} is as small as -8 MHz within the 13-ns 10%-intensity window of the OPO pulse.

Preliminary studies have been made of other experimental parameters that influence Δf_{inst}. The chirp is found to vary linearly as the OPO cavity resonance frequency is detuned away from the TDL seeder frequency, by alteration of the piezoelectric translator's locking voltage; typically, Δf_{inst} changes by $+7.5$ MHz if the OPO cavity is detuned by $+100$ MHz relative to the seed frequency—this is effectively a cavity-pulling process. The magnitude of Δf_{inst} also increases with pump-pulse energy, consistent with phase perturbations arising from nonlinear processes.

These measurements show that OPO frequency chirp depends on several experimental parameters: seed laser detuning (i.e., phase mismatch Δk), cavity resonance detuning (i.e., cavity pulling), and pump-laser energy. Small shot-to-shot fluctuations in these parameters may affect the stability of the OPO signal output frequency. In this context, Fig. 4 shows a representative set of seven separate measurements with the TDL-seeded OPO signal output wavelength set 0.08 nm below λ_{free} and pumped with pump-laser fluence set at ~ 70 μJ (twice the free-running OPO threshold). The mean overall chirp, Δf_{inst}, under these conditions is -46 ± 5 MHz. Figure 4 does not show accompanying fluctuations of ± 10 MHz in the absolute OPO signal frequency; these are attributable to small pulse-to-pulse changes in OPO resonator length, independent of chirp minimization, and could be improved by better cavity stabilization.

In practice, one can maintain Δf_{inst} as low as 10 MHz (including shot-to-shot variations) by minimizing the pump energy and adjusting the crystal temperature to match λ_{free} to the injection-seeded OPO wavelength. This chirp is significantly less than the observed Fourier-transform-limited optical bandwidth (60 MHz FWHM).

In conclusion, we believe that we have for the first time characterized the frequency stability and chirp of a nanosecond-pulsed, injection-seeded OPO to attain reliable spectroscopic control in high-resolution applications. Previous experiments and modeling on a birefringently phase-matched KTP OPO[10,11] studied temporally averaged frequency shifts, but our work on a PPKTP OPO now characterizes dynamic frequency perturbations during each OPO pulse. Importantly, one can control the magnitude and sign of the frequency chirp by adjusting λ_{free} relative to the wavelength of the injection seeder. The chirp is minimized when these two wavelengths coincide, which corresponds to minimal phase mismatch Δk. The smallest overall chirp measured was $\Delta f_{inst} \approx -8$ MHz for an OPO signal wavelength of 841.52 nm (0.04 nm below λ_{free}). We aim to extend these investigations to characterize frequency chirp in a higher-power OPO–optical parametric amplifier system and thereby design narrowband chirp-compensated systems for high-resolution pulsed spectroscopy. We shall also use a custom-built SLM Nd:YAG pump laser with a longer pulse duration (~ 30 ns FWHM) to reduce the Fourier-transform bandwidth of the OPO output and further improve the frequency-chirp characteristics.

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