Demonstration of a frequency-modulated, pulsed optical parametric oscillator

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We demonstrate that injection seeding a pulsed optical parametric oscillator with frequency modulated cw light with a modulation period equal to the cavity round-trip time produces pulses that have the same modulated character as the seed. A sensitivity of 10^{-3} was demonstrated for these pulses in frequency-modulated absorption measurements.
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Frequency-modulation (FM) of cw laser light has proven useful in sensitive absorption measurements, attaining shot-noise-limited performance[1,2] with absorption sensitivities as low as 10^{-8}. A lower sensitivity of 10^{-2} has also been demonstrated by frequency modulating nanosecond dye laser pulses.[3] Recently, Eyler *et al.*[4] demonstrated a sensitivity of 10^{-4} by pulse amplifying frequency-modulated cw light in a chain of dye amplifiers. As an alternative to dye lasers, nanosecond frequency-modulated optical parametric oscillators (OPOs) would be attractive because they should achieve sensitivities comparable to the dye lasers but with broader tunability, particularly in the infrared.

There are several ways an OPO could be used to produce FM light. The most obvious is to modulate the output light from a single-mode oscillator. Unfortunately, most high-frequency modulators have rather low optical damage limits. Another method is to use an FM pulse to pump a singly resonant OPO that is seeded at the signal wavelength. The resonated signal wave would remain single frequency, but the unresonated idler would be frequency modulated because the modulation phases must satisfy $\phi_{\text{pump}}=\phi_{\text{signal}}+\phi_{\text{idler}}$. The phase of the idler wave would mimic that of the pump. Such FM pulses could be created by modulating the light emitted by the pump laser, or by seeding the pump laser with an FM seed. Multiple-mode seeding has been previously demonstrated for a Nd:YAG laser.[5] A third method, and the one of interest here, is to seed a singly resonant OPO with an FM signal wave that has a modulation period equal to the round-trip time of the OPO cavity. Matching the modulation period to the cavity round-trip time allows the carrier and sidebands of the FM seed to coincide in frequency with adjacent OPO longitudinal cavity modes and be simultaneously amplified.

We showed in an earlier paper that a pulsed OPO tends to produce FM light,[6] and that it can produce a pulse with spectral properties that mimic the seed if the seed is FM. It will not mimic the seed if it has substantial amplitude modulation (AM), for example if the OPO were seeded on only two longitudinal modes. Viewed in the time domain, modulating the seed at the frequency of the OPO mode spacing ensures that the phase of the signal light after one traversal of the cavity is the same as that of the seed light incident on the input mirror. If we follow any point on the signal wave as it reflects around the cavity, we cannot distinguish FM seeding from monochromatic seeding. All points of the signal wave have the same field amplitude and are in phase with the seed light after one round trip of the OPO cavity. This equivalence of FM and monochromatic seeding depends on group velocity dispersion effects being insignificant, or equivalently, on the acceptance bandwidth of the crystal being much larger than the modulation frequency. Typical acceptance bandwidths are several hundred GHz compared with practical modulation frequencies of less than 10 GHz, so this condition should be met for most OPOs. In the FM-seeded OPO the idler wave assumes a modulation phase opposite that of the signal. The idler could also be resonated, but this makes continuous tuning of the OPO difficult. Of course, the three methods described above for creating FM light with OPOs could be used in combination to produce frequency modulation at a variety of frequencies, but we will consider only the FM-seeded method in the remainder of this letter.

The FM absorption measurement technique relies on the conversion of frequency modulation to AM near sharp absorption features, so one key to high absorption sensitivity using FM techniques is a sufficiently large modulation index combined with strongly suppressed AM. Any residual AM adds an unwanted background to the absorption signal. The key question for our study then is whether the frequency modulation of the seed is reproduced in the OPO light, and how well the AM is suppressed. As we will document here, our FM-seeded OPO produces FM light of sufficient quality to achieve absorption sensitivities of at least 10^{-3}. Because both the signal and idler waves are frequency modulated, either can be used for spectroscopy. As a practical matter, the waves can be sum-frequency mixed after the spectroscopic sample to create light at the pump wavelength that has the same AM character as the spectroscopic beam. Thus infrared FM spectroscopy can be implemented with detection at the fixed pump wavelength where high-speed detectors are readily available.

Figure 1 shows a diagram of our experiment. A frequency-doubled spatially filtered single-longitudinal-mode Nd:YAG laser provides 7 ns duration pulses to pump a KTP ring OPO operating at a signal wavelength of 800 nm and an idler wavelength of 1588 nm. With a pump beam diameter of 1 mm at the $1/e^2$ irradiance points, the threshold for oscilla-

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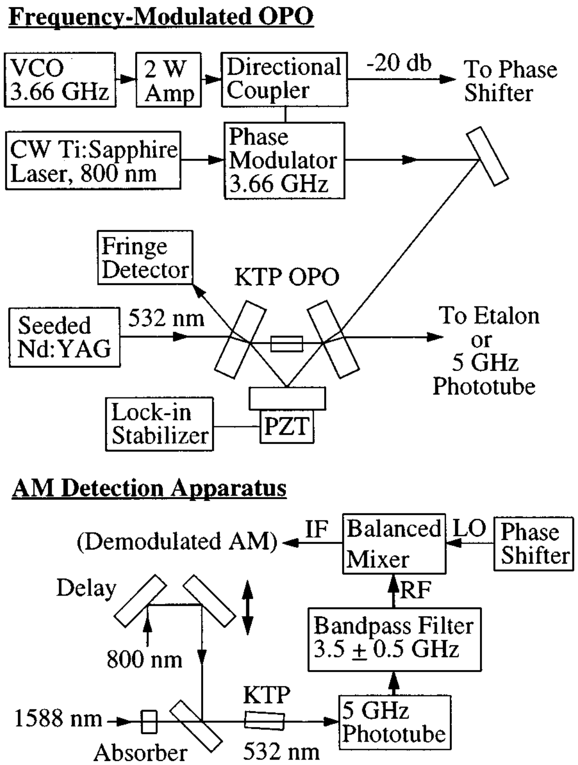

FIG. 1. Diagram of the FM-seeded OPO and AM detection apparatus. (VCO) voltage-controlled oscillator.

tion is ~1.2 mJ. Light from a cw Ti:sapphire laser is frequency modulated at 3.66 GHz, the free spectral range of the OPO cavity to serve as the signal seed. The frequency modulation index is typically 0.75, meaning the ratio of the power in each of the first FM side bands to the carrier power is 16%. The optical power in the seed beam is ~4 mW. One of the OPO mirrors is translated by a piezoelectric mount to lock the OPO cavity resonance to the seed light. A lock-in stabilizer maintains cavity resonance by dithering the OPO cavity length while monitoring the intracavity seed light intensity by detecting light leaking through one of the cavity mirrors. The signal light or signal-plus-idler sum-frequency light is detected using Hamamatsu phototubes with bandwidths of 5 GHz. Their output is directed to either a 4.5 GHz transient digitizer (Tektronix SCD5000; detector-plus-digitizer 3 db bandwidth of 3.2 GHz), or passed through an electronic bandpass filter to suppress the pulse envelope, with the remaining AM demodulated by mixing with the 3.66 GHz local oscillator in a balanced mixer. We separated the signal and idler waves after the OPO using beam splitters so we could make measurements on each beam individually, and recombined them with an adjustable time delay in a KTP crystal to create 532 nm sum-frequency light. The total energy contained in the signal and idler pulses is less than 2 mJ, and the sum-frequency conversion efficiency is typically 2%.

We first compared the spectra of the seed light and the OPO signal pulse using a scanning Fabry–Perot étalon. We find that if the OPO cavity length is adjusted to match the cavity free spectral range to within a few percent of the modulation frequency, and the crystal angle is adjusted to minimize phase mismatch Δk, the OPO spectrum nearly duplicates the seed spectrum, as shown in Fig. 2.

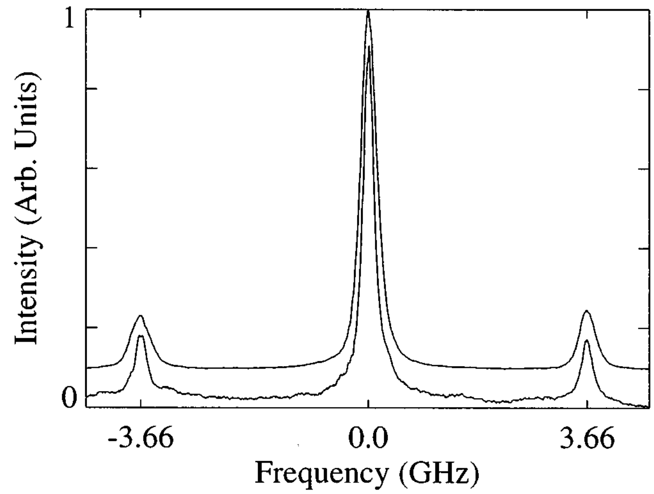

FIG. 2. Scanning Fabry–Perot étalon spectra of the FM seed (lower) and OPO signal (upper). Spectra are offset for clarity.

We next examined the signal time-profiles using the transient digitizer to determine whether AM is strongly suppressed in the OPO light. Figure 3(a) shows a typical OPO signal pulse with 1% or less AM, and Fig. 3(b) shows a similar signal pulse after passing through an AM inducing absorber. In the absence of the intentionally induced AM there was often AM with an amplitude of ~1% of the pulse amplitude, so while the AM was small it was not always totally suppressed. The small residual AM observed in Fig. 3(a) produced a strong demodulated AM signal using the detection apparatus shown in Fig. 1, and it suggests a slight imbalance in the amplitude of the FM sidebands, or phase distortion, introduced by the OPO. The most likely source of the imbalance is imperfect matching of the seed spectrum to the cavity spectrum resulting in AM on the seed circulating in the OPO cavity at the start of the amplification process. We found that we could reduce the AM by a factor of

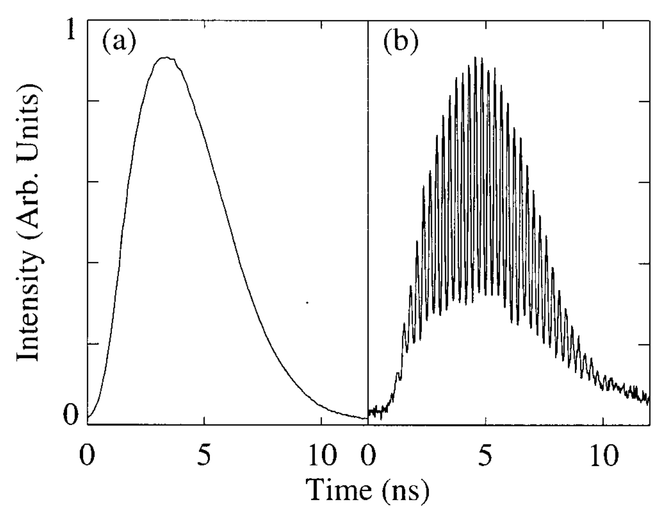

FIG. 3. OPO signal pulses measured with a 3.2 GHz bandwidth detection system: (a) undisturbed pulse and (b) after passing through an absorption cell filled with room temperature rubidium vapor. The seed wavelength was tuned to the Rb $5S_{1/2} \rightarrow 5P_{3/2}$ resonance at 780 nm for this measurement.

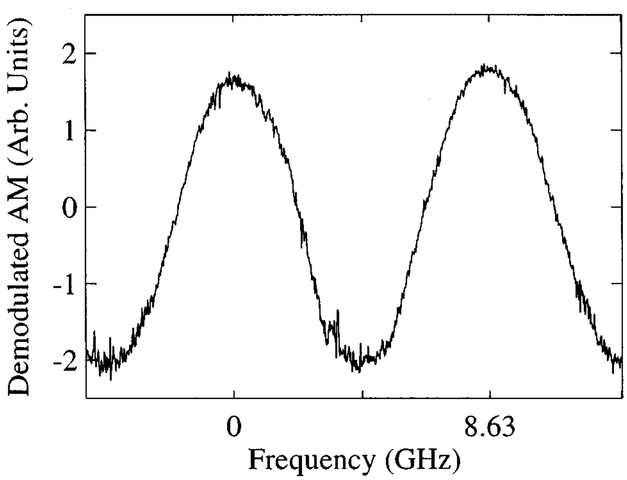

FIG. 4. Spectrum of demodulated AM normalized to the 532 nm signal-plus-idler sum-frequency-mixed pulse energy. The spectrum is generated by placing an 8.63 GHz free-spectral-range uncoated optical flat in the 1588 nm idler beam and scanning the 800 nm FM light that seeds the OPO.

$\sim$10 by modifying the cavity lock-point to be slightly away from the position that gave the highest circulating seed power. This strengthens one of the sidebands slightly and weakens the other resulting in improved balance and less AM. With this modified locking method, we tested the absorption sensitivity of our system by inserting in the 1588 nm idler beam an uncoated optical flat with a free spectral range of 8.63 GHz. Scanning the seed wavelength and monitoring the demodulated AM on the 532 nm sum-frequency light produces spectra like that shown in Fig. 4. In this scan we averaged 5 pulses at each point and normalized the AM signal by the 532 nm pulse energy. Combining the Fabry–Perot transmission function of the optical flat with the signal spectrum we calculate a maximum AM of 0.09 where

$$\mathrm{AM} \equiv \frac{P_{\max} - P_{\min}}{P_{\max} + P_{\min}} \tag{1}$$

and $P_{\max}$ and $P_{\min}$ are the light powers at the maximum and minimum of the amplitude modulation. This degree of AM is equal to that generated by absorption of about 20% of one sideband, so for calibration of our system we equate the maximum signal in this scan with a 20% absorption. From the signal-to-noise ratio of this scan, we conclude that we could measure absorptions as small as 10^{-3}. Although this sensitivity is low compared to typical cw FM spectroscopy absorption sensitivities, it should be compared with the Fourier component of our pulse envelope in the detection band, which is $\sim 10^{-6}$. Clearly, we have not yet reached this limit. Rather, the primary sensitivity-limiting noise source appears to be AM on the pump pulse, which is impressed on the signal and idler pulses. The measured peak-to-peak amplitude modulation of the pump is about 10^{-3}, indicating that longitudinal modes lying 3.66 GHz from the pump's seed frequency are suppressed to $\sim 10^{-6}$ of the seeded mode. Placing an étalon in the pump beam or in the pump laser cavity could further suppress these modes and perhaps lower the pump AM by a factor of 10, improving the absorption sensitivity of our system to 10^{-4}. Sensitivity could be further improved by measuring residual AM before the absorber, and subtracting it from the AM signal of interest.

As a final note, we find that when pumped several times above threshold, the OPO produces pulses with about 1% AM even when seeded by a signal wave with strong AM. The OPO tends to suppress the AM to this level even with no attempt to minimize the AM of the seed light circulating in the cavity.[6] Given the OPO's tendency to suppress AM, an absorption sensitivity of 1% should be possible with little effort.

In conclusion, we have shown that it is possible to generate high quality FM light from an OPO by properly seeding it with FM light. We demonstrated an absorption sensitivity of 10^{-3} and believe 10^{-4} should be possible by averaging over more pulses and by using a pump source with less AM.

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