

All-solid-state, tunable, single-frequency source of yellow light for high-resolution spectroscopy

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We demonstrate a cw doubly resonant optical parametric oscillator that is frequency doubled in an external resonant cavity to the visible spectral range. We obtained single-frequency radiation in the range 565–590 nm with as much as 3.8 mW of power, which is continuously tunable over an 18-GHz range and step tunable over 160 GHz. The source is well suited for high-resolution spectroscopy in the visible region. As a demonstration, we performed persistent hyperfine spectral hole-burning spectroscopy of $\text{Eu}^{3+}:\text{Y}_2\text{SiO}_5$. Reliable operation of the source permitted studies of the hole's lifetime over several hours. © 2001 Optical Society of America
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The replacement of ubiquitous and time-proven cw dye lasers by all-solid-state sources is an outstanding challenge in laser development. In recent years the evolution of frequency-stable cw optical parametric oscillators (OPOs) as sources of tunable, coherent radiation has progressed rapidly^{1,2} and thus these sources may be considered a possible alternative to dye lasers. It has been shown that their wide tuning range, good frequency stability, and high efficiency make them well suited for high-resolution spectroscopy in the near and midinfrared.^{3–5}

Obtaining access to the visible spectral region with a cw OPO is, however, more challenging because of the requirements on materials and pump sources. One approach is to use a UV pump source.⁶ All-solid-state, single-frequency UV lasers with good frequency stability are feasible but expensive. In addition, attainment of a UV-pumped OPO based on currently available nonlinear crystals will be hampered by problems such as significant absorption of the pump wave, low nonlinearity, and walk-off effects.

An alternative approach to obtaining access to the visible is frequency conversion of the output of a near-infrared OPO, building on the benefits of easily available all-solid-state pump sources and utilizing common available crystals with good nonlinearities.⁷ In particular, diode-pumped, frequency-doubled Nd:YAG lasers with single-frequency output make good pump sources. In this Letter we describe what we believe to be the first cw frequency-doubled OPO operating in the visible and its application to high-resolution spectroscopy of rare-earth ions in a crystal.

We chose a doubly resonant OPO (DRO) design in which signal and idler waves are resonant. Resonating both the signal and idler wavelengths has two advantages: First, double resonance gives a low threshold ($\ll 100$ mW), and second, it permits smooth tuning of the output frequencies over a usefully large frequency interval by tuning of the pump frequency. Among OPOs, DROs are highly sensitive to disturbances and therefore often are viewed as impractical for applications. However, with an appropriate cavity-length control system, high stability can be achieved with operation on a single mode pair without mode hops in excess of 10 h.^{8,9} As a consequence of this stability, it is feasible to convert the OPO

output in an additional stage, for example, by use of a resonant frequency doubler. It thus becomes possible to generate tunable single-frequency visible light at useful power levels from an all-solid-state source.

Our single-frequency source of yellow light is composed of three nonlinear stages (Fig. 1). In the first stage a commercially available monolithic ring Nd:YAG laser, producing 1.05 W of power at 1064 nm, was used to pump a semimonolithic resonant frequency doubler. This doubler contains a bulk $\text{MgO}:\text{LiNbO}_3$ crystal and was described previously.¹⁰ Currently it generates as much as 550 mW of power at 532 nm.

The second stage consists of a DRO that utilizes the same design and crystal as the doubler. For the output wavelength range of interest (near 1160 nm) the crystal was kept near 120 °C by use of an oven with 2-mK stability. The output coupler ($r = -25$ mm) has 98% reflectivity at both the idler and the signal wavelengths and 95% transmission for the pump and was mounted upon a piezoelectric transducer (PZT) actuator for feedback control. The cavity mirror was formed with a high-reflection coating on the curved ($r = -10$ -mm) back surface of the nonlinear crystal. This coating is also highly reflective at the pump wavelength. The short cavity has a free spectral range

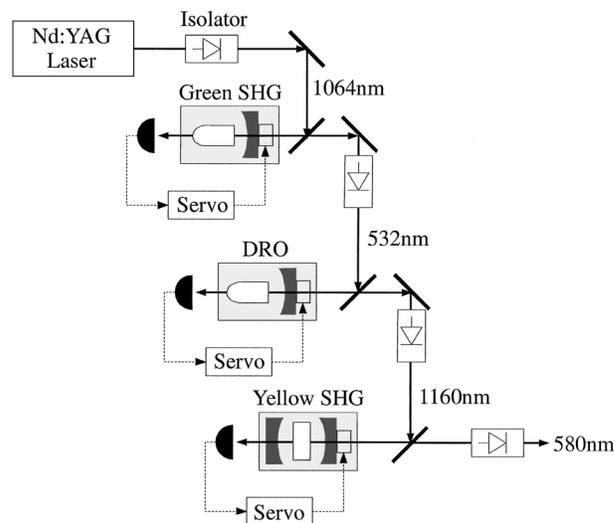


Fig. 1. Schematic of the all-solid-state yellow source.

of 4.2 GHz. Round-trip cavity loss was 1%. Good mode matching (60%) and low loss yielded a pump threshold of 15 mW at 532 nm, and, for a pump power of 450 mW, as much as 95 mW of power is generated at the idler wavelength. Including the signal power, the power-conversion efficiency was 46%. Changing the MgO:LiNbO₃ phase-matching temperature tunes the idler wavelength from 1130 to 1190 nm.

The third nonlinear stage is another resonant second-harmonic generator, in which the idler wave of the DRO is doubled into the yellow spectral region. The nonlinear crystal is a periodically poled lithium niobate multigrating (8 mm wide, 0.5 mm thick, 16 mm long) with seven gratings (periods 8.0–8.6 μm , each 1 mm wide), which are suitable for second-harmonic generation (SHG) of the full spectral range of the DRO output. Operation with the 8.6- μm grating at 140 °C was chosen to prevent photorefractive effects. A dual-layer oven that uses both heating rods and high-temperature Peltier elements provided millikelvin temperature stability.

The periodically poled lithium niobate crystal is antireflection coated on both ends. The input–output coupler mirror ($r = -25$ mm) has $R = 98\%$ at 1160 nm and $R < 4\%$ at 580 nm and is mounted upon a PZT actuator; the back mirror ($r = -30$ mm) is highly reflective at 580 and 1160 nm. The cavity finesse was measured to be 220. For the 28 mW of power that is available at 1160 nm and with 66% mode matching, as much as 3.8 mW of yellow light was obtained that was tunable from 565 to 595 nm. Because each of the three cavities used is of the standing-wave type, 40-dB optical isolators were placed between the cavities. The isolator between the OPO and the second SHG stage had significant loss at 1160 nm, with the result that only 60% of the generated OPO light was incident upon the doubling cavity. The signal wave is not currently used; however, with modification of the optics it would be possible to frequency double the wave in the same setup to give visible light in the 483–502-nm region.

Resonance of each cavity was maintained with a feedback-control system based on the FM technique.⁸ Sidebands generated by application of a radio-frequency voltage transversely to the MgO:LiNbO₃ crystal in the first doubler provided the error signal for both the first doubler and the DRO, whereas an external electro-optic modulator between the DRO and the second doubler provided the sidebands for locking the doubler. To enhance stability, each resonant cavity was also isolated from mechanical and acoustic noise and air temperature fluctuations by appropriate shielding. As a result, the DRO operated continuously without mode hops for extended periods, as illustrated in Fig. 2. Starting with 0.1% intensity noise for the 1064-nm laser output, the power noise remained low after conversion, with 0.5% at 532 nm, 1% at 1160 nm, and 3% at 580 nm.

Tuning the yellow light to a particular frequency was accomplished in several steps. Coarse tuning was done by adjustment of the OPO crystal temperature, with an average tuning coefficient of -3 THz/K for the yellow light. As a result of heating of the OPO crystal

by the pump, signal, and idler waves, the OPO also exhibited frequency sensitivity to pump power, with an average coefficient of 9 GHz/mW of pump power.

We found that, by unlocking the DRO cavity when it was locked to a particular signal–idler mode, adjusting the dc offset voltage to the PZT actuator, and relocking the cavity, it was possible to step sequentially through the mode pairs of the cavity, similar to the approach described in Ref. 3. Figure 3 illustrates a series of 20 of these operations, covering a range of approximately 80 GHz at 1160 nm.

Fine tuning without mode hops is achieved by tuning of the Nd:YAG laser frequency. This is possible in either of two ways: by fast tuning over a 200-MHz range by application of a voltage to a PZT actuator mounted upon the Nd:YAG crystal or by slow tuning over the free spectral range of the monolithic laser cavity (10 GHz) by a change in the temperature of the laser crystal. Using the latter yielded a maximum continuous tuning range of 18 GHz at 580 nm, limited only by the free spectral range of the pump laser. It can be estimated that a continuous tuning range of ~ 100 GHz should be possible with an appropriate pump source.⁸ Because the frequency separation between OPO cavity modes is less than the tuning range of the pump laser, by a combination of

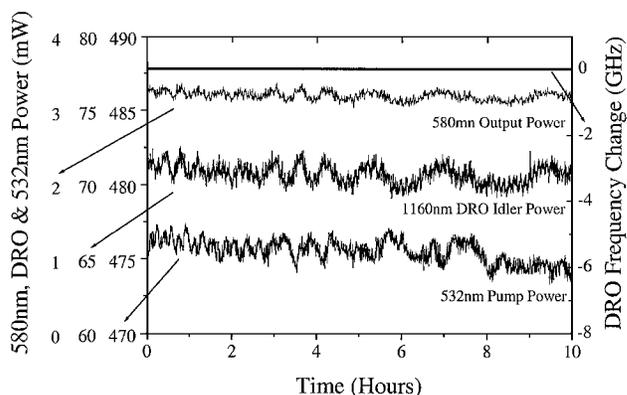


Fig. 2. Power and frequency stability of the DRO over a 10-h period. The resolution (100 MHz) of the wavemeter is less than the laser frequency drift over this period but greater than the intermode spacing (4.2 GHz).

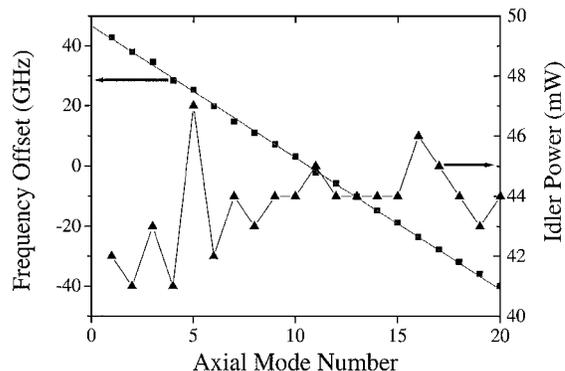


Fig. 3. Sequential stepping of the DRO axial mode in small incremental dc offsets on the DRO cavity PZT. As many as 20 adjacent modes can be locked in this manner, giving a smooth tuning range of 80 GHz at 1160 nm.

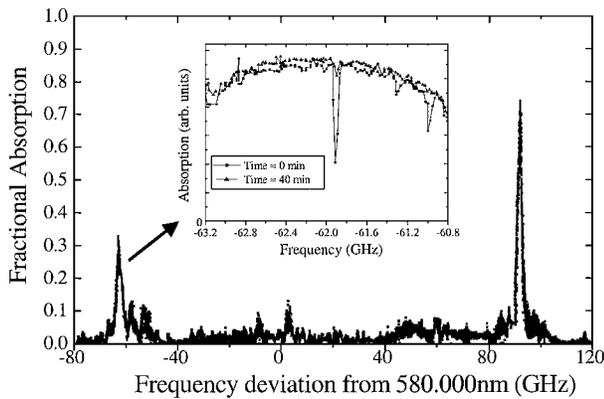


Fig. 4. OPO spectroscopy of the ${}^5D_0 \rightarrow {}^7F_0$ transition of $\text{Eu}^{3+}:\text{Y}_2\text{SiO}_5$ at 4 K. Inset, resultant spectral holes and antiholes burned into the center of the inhomogeneously broadened transition of site 1 from a single-second-long write beam.

controlled mode jumps and laser tuning a complete frequency coverage can be achieved over a large frequency interval. At 580 nm, a range of as much as 160 GHz is coverable in this way. Coverage of this extent is useful and sufficient for many spectroscopic studies. The primary source of frequency instability of the DRO is the frequency drift of the pump laser. Because of the very low pump laser frequency instability (below 30 MHz/h), the frequency drift at 580 nm was typically less than 55 MHz/h, determined by measurements with a wavemeter.

Using this laser source at 580 nm, we examined the spectroscopy of the ${}^5D_0 \rightarrow {}^7F_0$ transition of $\text{Eu}^{3+}:\text{Y}_2\text{SiO}_5$ at cryogenic temperatures. This transition is of interest because it exhibits the lowest known homogeneous linewidth for an optical transition in a solid.¹¹ An uncoated 5 mm \times 5 mm \times 10 mm Y_2SiO_5 crystal doped with 0.1% Eu^{3+} , cooled in a liquid-He cryostat, was used. We used a wavemeter to accurately monitor the DRO frequency. Tuning the frequency-doubled output of the DRO in the manner described above, we obtained the spectrum of the transition (Fig. 4), which clearly exhibits two peaks that correspond to the two Eu^{3+} sites within the Y_2SiO_5 unit cell. Significant peak absorption (site 1, 34%; site 2, 75%) was detected both by measurement of absorption and of fluorescence. The inhomogeneously broadened spectrum of each site (site 1, $\lambda = 580.070$ nm, $\Delta\nu = 3$ GHz; site 2, $\lambda = 580.244$ nm, $\Delta\nu = 2.5$ GHz) could be scanned continuously without DRO mode hop. We performed persistent spectral hole burning at 4 K, with typical burn durations of 1 s. The inset of Fig. 4 shows a hole spectrum recorded 40 min after burning (linewidth, <1 MHz). By recording the hole spectrum at intervals of a few hours we could follow the hole decay as long as 15 h after burning, limited by the intensity required for reading out the spectral information.

In summary, we have described a widely tunable, all-solid-state visible light source that is well suited

for high-resolution spectroscopy. As much as 3.8 mW of single-frequency light was generated in the range 565–590 nm, which can be tuned continuously over 18 GHz and can be sequentially stepped over a range of 160 GHz. Single-mode operation has been observed for as long as 10 h without stabilization to a frequency reference. The usefulness of the source is demonstrated through spectroscopy of the ${}^5D_0 \rightarrow {}^7F_0$ transition of $\text{Eu}^{3+}:\text{Y}_2\text{SiO}_5$ at 4 K, where the inhomogeneously broadened two-site spectrum as well as persistent hole burning was observed. By measuring the hole's lifetime, we showed that tuning a DRO repeatedly to the same frequency over the course of more than a dozen hours is possible.

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