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Frequency stabilization and high resolution spectroscopy using frequency-doubled Nd:YAG lasers

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Monolithic diode-pumped solid state lasers [1] are compact, reliable and efficient light sources, with inherently narrow linewidth and low intensity noise. These features are very attractive for applications that require high spectral purity - e.g., optical frequency and length standards, metrology and optical coherent communication. However, realization of these applications has been plagued by the lack of suitable absorbers for absolute frequency stabilization within the relatively narrow tuning range of the lasers. For example, although CO_2 [2] and Cs_2 [3] have several absorption lines within the tuning range of the 1064 nm Nd:YAG laser, these lines are extremely weak and require relatively high pressure or temperature to obtain sufficient absorption for frequency locking. [3]

An alternative approach is second harmonic generation and locking to absorption lines in the visible. [4] Unfortunately, conventional single pass second harmonic generation of low power CW lasers is highly inefficient, e.g., frequency doubling of a 100 mW 1064 nm Nd:YAG laser in a 10 mm MgO:$LiNbO_3$ yields only 23 μW of green light. Here we demonstrate two techniques to improve the conversion efficiency - resonant doubling [5] of the 1064 nm Nd:YAG laser and single pass quasi-phasematched waveguide doubling [6] of the 1319 nm Nd:YAG laser. The second harmonic light is used to study the $^{127}I_2$ absorption lines and to absolutely stabilize the laser frequency.

Two 1064 nm Nd:YAG lasers have been frequency doubled using monolithic, MgO:$LiNbO_3$ resonant doublers. The pump beam followed a triangular path in each doubler, bounded by two total internal reflections and a reflection from the dielectrically coated input coupler. The resonant enhancement of the fundamental beam significantly improved the conversion efficiency - up to 159 mW at 532 nm were generated with 300 mW of 1064 nm radiation. Frequency tunability with a fixed output power was achieved by fringe side locking of the resonant doubling crystal to the laser frequency through a servo which controls the crystal temperature. [7]

The 532 nm output was the source for FM Doppler-free spectroscopy of $^{127}I_2$ in the range 18787-18789 cm^{-1}. [8] We used 8.5 and 15 cm long iodine cells, held at a temperature of 0°C (pressure ~ 30 mtorr). We have observed 8 strong ro-vibrational lines, between the lowest vibrational level in the X state to the vibrational levels 32-36 in the B state. Several additional transitions between the first and second vibrational levels in the ground state and the vibrational levels 35-42 in the B state have also been detected. Fig. 1 shows the Doppler-free FM spectrum of R(86)33-0 line, with a calculated center frequency [9] of 18787.2792 cm^{-1} (Line # 1107 in iodine atlas [10] with a measured center frequency of 18787.2800 cm^{-1}).

The FM signal of each hyperfine component can be used as an error signal to lock the laser frequency. To measure the stability we built two independent, nearly identical systems, and locked each one of the two lasers to the a_1 line of R(56)32-0 at 18788.3 cm^{-1} (Line # 1110 in Ref. [10]). For integration time $\tau > 2$ ms, the two sample deviation of the beat frequency between the two lasers (root Allan variance[11]) follows a $1.1 * 10^{-12}/\sqrt{\tau}$ dependence, and the lowest value of $2.5 * 10^{-13}$ (frequency deviation of 70 Hz) is reached at $\tau = 32$ s. The maximum excursion of the beat frequency over a 1 hour period is approximately 2 kHz. [8]

We have measured the splitting of six iodine ro-vibrational lines by heterodyne spectroscopy. [8] The measured spectra were used to determine the hyperfine constants for these transitions. Following the procedure outlined by Foth and Spieweck, [12] the Hamiltonian of the hyperfine interactions can be written as:

$$H_{hfs}=H_{EQ}+H_{SR}+H_{SSS}+H_{TSS}$$

where H_{EQ} (eQq), H_{SR} (C), H_{SSS} (a), H_{TSS} (d) represent the electric quadruple, spin-rotation, scalar spin-spin and tensor spin-spin interactions, respectively. The symbols in parenthesis denote the constants of each of these interactions. For the electric quadruple interactions we have also considered rotational levels separated by ±2. The frequency splitting depends strongly on the *difference* in the hyperfine constants between the B and X states, but exhibits only weak dependence on the *absolute* values of these constants. We have therefore used fixed values for the constants of the X state, [13] while fitting the parameters of the B level to the experimental measurements.

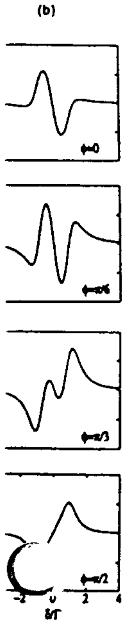

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Table 1 summarizes the obtained fitting constants and standard deviation for the six measured lines. The standard deviation of the fit for all lines is better than 10 kHz. There are two pairs of lines belonging to the same vibrational bands (32-0, 33-0). The hyperfine components depend primarily on the vibrational level (only eQq exhibits a weak dependence on the rotational number). Indeed, the independently fitted values of the hyperfine constants for these two pairs are in good agreement. The dependence of the ΔeQq and ΔC constants on the vibrational numbers is consistent with measurements of iodine lines at other wavelengths [14]: While only slight changes are observed in ΔeQq, ΔC increases by 50% as the vibrational level of the B state increases and approaches the dissociation limit.

Table 1: Standard deviation of the fit (σ) and hyperfine constants difference.

Line	σ (kHz)	ΔeQq (MHz)	ΔC (kHz)	Δa (kHz)	Δd (kHz)
P(53)32-0	6.51	1908.4757±0.08	86.047±0.15	-10.27±4.4	-44.4±3.7
R(56)32-0	6.92	1908.4057±0.1	86.34±0.23	-10.60±5.4	-44.95±4.5
P(83)33-0	4.50	1906.9447±0.077	94.483±0.56	-10.14±4.2	-48.65±4.5
R(86)33-0	2.25	1906.8107±0.044	95.043±0.05	-10.09±1.4	-48.54±0.7
R(106)34-0	7.13	1905.2577±0.125	104.829±0.18	-9.87±5.7	-53.67±5.4
R(134)36-0	9.81	1902.2662±0.15	128.694±1.6	-15.14±8.6	-64.7±7

Second harmonic generation and locking to I_2 transitions in the visible was also performed using the 1319 nm Nd:YAG laser. This laser operates near the 1.3 μm transmission window in optical fibers. Hence, absolute frequency stabilization may be used for frequency references in densely packed wavelength-multiplexed or coherent fiber-optic communication systems, [15] for 'cold start' coherent communication and for fiber-optic sensing applications. The second harmonic was generated by single pass doubling in a quasi-phasematched waveguide. Quasi-phasematching in $LiNbO_3$ is accomplished by periodic ferroelectric domain reversal, [6] and allows the use of the large d_{33} nonlinear coefficient which is not accessible to birefringent phase matching. The improvement in conversion efficiency compared to the birefringent process is $(2d_{33}/\pi d_{31})^2 \sim 20$. Further improvement in conversion efficiency is obtained by the waveguide confinement. The waveguide doubler was fabricated by the formation of a Ti indiffused ferroelectric domain grating with a 10.5 μm period, [16] followed by annealed proton exchange channel waveguide. Phase matching was obtained at ~35 °C. With 100 mW pump coupled into the 5 mm long waveguide, a ~1 mW of red light was generated, exceeding the birefringently phasematched bulk conversion efficiency by a factor of 170.

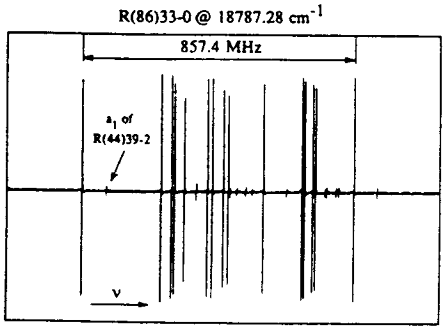

Fig. 1: FM saturated absorption spectrum of R(86)33-0 @ 18787.28 cm^{-1}. The weak lines in this spectrum belong to the R(44)39-2 transition. Modulation frequency is 4 MHz, cell length = 8.5 cm and cell temperature is 0 °C.

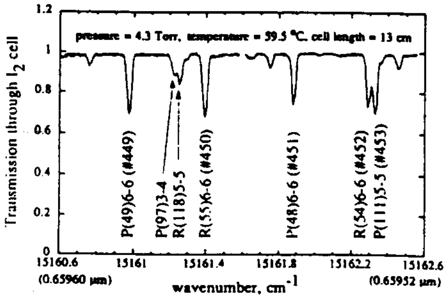

Fig. 2: I_2 absorption lines within the tuning range of the frequency-doubled 1319 nm Nd:YAG laser. Line numbers in parenthesis are taken from Ref. [10].

The I_2 transitions at 659 nm are much weaker than at 532 nm, hence we have heated the 13 cm long iodine cell to temperature between 40-70 °C to increase the absorption. Fig. 2 shows the 10 observed absorption lines within the tuning range of the doubled laser, and the assignment of the stronger transitions. The Doppler-broadened P(48)6-6 transition (linewidth ~800 MHz), with a calculated center frequency of 15161.8796 cm^{-1} (Line # 451 in Ref. [10], measured frequency 15161.8813 cm^{-1}) was used as a frequency reference. We used FM spectroscopy to obtain an error signal: The second harmonic was frequency modulated at 105 MHz and passed through the I_2 cell held at 57 °C (pressure ~ 3.5 Torr). The detected signal was mixed with a local oscillator, and fed through a servo amplifier to the piezo frequency actuator of the laser. The frequency stability was evaluated using an independent I_2 cell. We measured the frequency variation over 16.5 minutes and obtained for an integration time of 0.1 s a standard deviation of 210 kHz, an Allan variance of $2.4 * 10^{-10}$, and a maximum frequency excursion of 1.06 MHz. This simple technique was sufficient to eliminate the typical ~MHz per minute drift rate of the laser, and to stabilize the laser at a wavelength of 1319.098 nm. A tenfold improvement in conversion efficiency may be obtained by increasing the waveguide length and optimizing its dimensions. This will enable locking to Doppler-free lines, as well as extending the technique to lower power semiconductor lasers.

The combination of inherently stable monolithic diode-pumped solid-state lasers and efficient techniques for frequency doubling enable new spectroscopic and metrological applications. The I_2-stabilized 1064 nm Nd:YAG laser offers important advantages with respect to other frequency standards in the visible - higher power and stronger I_2 transitions with respect to the I_2-stabilized He-Ne at 633 and 612 nm; narrower linewidth and smaller, more efficient laser than the I_2-stabilized Ar^+ laser. The 1319 nm Nd:YAG may be used as a frequency or wavelength reference in fiber optic communication and sensing systems. In addition, the availability of two harmonically related wavelengths is advantageous for precision length measurements in dispersive media, e.g. in air.

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