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Observation of iodine transitions using the second and third harmonics of a 1.5- μm laser

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ABSTRACT This paper presents spectroscopic measurements of iodine at 778 nm and 518 nm performed by second and third harmonics of a 1.5- μm diode laser, generated by quasi-phase matching in a periodically poled lithium niobate waveguide. Sub-Doppler spectroscopy by a 780-nm source was also demonstrated, and shows the potential of the system to reach high levels of frequency stability by locking the laser to the iodine transitions. The suggested method significantly improves the number of frequency references available for stabilizing 1550-nm lasers.

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1 Introduction

Optical frequency standards and references have been extensively used, both in fundamental research, such as physical constants definition [1, 2], and as an essential tool for novel technologies and applications. Since the importance of 1550-nm sources has grown over the last two decades as a result of their rising popularity in the telecommunication market, control and characterization of these sources has been intensively studied. Optical references in the C-band window (1529–1561 nm) can be utilized in dense wavelength division multiplexing systems and components, fiber-optics strain sensors [3] and other precision-measurement instrumentation. Basic performance can be obtained by temperature-controlled fiber Bragg gratings, which are employed in sensitive commercial systems. As more demanding applications arise, references utilizing absorption spectra of atoms and molecules supply higher-stability performance. Among absorption references at 1550 nm are found the molecules NH_3 [4], HCN [5] and C_2H_2 [6], which present absorption lines with a limited range, and the Kr and ^{87}Rb atoms, which require additional excitation to produce significant absorption and present only a few absorption lines.

Utilizing a wider range of absorption references is possible by non-linear frequency-conversion interactions, in

which the most commonly used one is second-harmonic generation (SHG). Studies using SHG by the ^{39}K [7, 8] and ^{87}Rb atoms have been demonstrated and have shown high levels of stability, but still limit the locking frequency choice to a small number of atomic transitions.

The $^{127}\text{I}_2$ molecule has been frequently chosen as a frequency reference since the upper and ground vibrational states $B^3\Pi_{ou}^+ \leftarrow X^1\Sigma_g^+$ provide a rich absorption spectrum containing approximately 100 000 vibrational allowed transitions spread over the 500–900-nm range. The iodine Doppler-broadened molecular absorption lines present a spectral FWHM of about 900 MHz and can be identified and predicted using the Gerstenkorn et al. I_2 atlas of absorption [9]. Higher levels of frequency stabilization are achieved by sub-Doppler techniques such as saturation spectroscopy and polarization spectroscopy, which enable stabilization to the hyperfine splittings with a natural spectral width at the MHz level. These techniques are supported by a recent publication [10], which improves the accuracy and availability of references based on heated iodine cells in the near infrared.

Previous work [11] demonstrated frequency stabilization of a 1319-nm Nd:YAG laser by generating a second harmonic and locking it to the absorption lines of an I_2 molecule. As for 1550-nm sources, no experimental demonstration has yet been published. In this paper, we present for the first time, to our knowledge, a spectroscopic result of the I_2 molecule obtained by SHG and third-harmonic generation (THG) using an amplified 1556-nm external cavity tunable diode laser as a source. The frequency conversion was performed by the quasi-phase-matching (QPM) method [12] in a highly efficient periodically poled lithium niobate (PPLN) waveguide device.

A special heated absorption cell was constructed for the study by the second-harmonic light at 778 nm, which enabled us to reach high temperatures that are required for the significant population of the ground state, while gas pressure in the cell is controlled to avoid pressure broadening. THG in the periodic waveguide was studied, and iodine spectroscopy at 518 nm by the third harmonic was also demonstrated.

In Sect. 2 SHG and THG results in the PPLN waveguide are presented, and in Sect. 3 we show spectroscopic measurements obtained by second and third harmonics. The main results are discussed and summarized in Sect. 4.

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2 Second- and third-harmonic generation

2.1 Second-harmonic generation

Annealed proton-exchange (APE) waveguides have been deployed in PPLN crystals as they enhance the non-linear frequency-conversion efficiency by tightly confining the optical beam and thus avoiding the conventional tradeoff between tight focus and effective length in a bulk configuration. Conversion efficiency in a waveguide is greater than in the bulk case by a factor of $\lambda_\omega L / 2n_\omega A_{\text{eff}}$, where A_{eff} is the effective area of the beam traveling through the waveguide, λ_ω is the fundamental wavelength and L is the interaction length.

SHG was obtained by coupling a New Focus 1550-nm external cavity tunable diode laser, followed by an erbium-doped fiber amplifier (EDFA) with 16 dBm of output power, into the PPLN waveguide. To avoid photo-refractive damage, the crystal was kept at a temperature of 87 °C by an electronic controller. The QPM period used was 14.5 μm and the phase-matching wavelength was 1556.2 nm, measured by a Burleigh wavemeter. Due to the waveguide's enhancement in efficiency of about two orders of magnitude compared to a bulk configuration, and the use of an optical amplifier, we were able to measure a second-harmonic power of 2 mW, which corresponds to a conversion efficiency of more than 125%/W of the total IR input power.

2.2 Third-harmonic generation

THG through $\chi^{(2)}$ interactions requires two processes to be phase-matched. SHG should be performed, and sum-frequency generation (SFG) between the second harmonic and the fundamental wave must also occur to produce the third harmonic. Equations (1a) and (1b) define the phase mismatch ΔK for both SHG and SFG:

$$\Delta K_{\text{SHG}} = \frac{4\pi}{\lambda_\omega} (n_{2\omega} - n_\omega), \quad (1a)$$

$$\Delta K_{\text{SFG}} = \frac{2\pi}{\lambda_\omega} (3n_{3\omega} - 2n_{2\omega} - n_\omega), \quad (1b)$$

where n_ω , $n_{2\omega}$ and $n_{3\omega}$ are the refractive indices for the fundamental, second and third harmonics, and λ_ω is the fundamental wavelength.

Efficient $\chi^{(2)}$ interactions require $\Delta k \ll L$, where L is the interaction length. One method of achieving the above condition is by modulating the non-linear coefficient in a periodic structure with a period Λ , thus producing the k vectors shown in (2):

$$K_m = \frac{2\pi m}{\Lambda}, \quad (2)$$

which creates an efficiency function η that is proportional to the expression in (3):

$$\eta \propto \text{sinc}^2 \left(\frac{\Delta k_{\text{QPM}} L}{2} \right), \quad (3)$$

where $\Delta k_{\text{QPM}} = \Delta k - k_m$.

As the period Λ can be chosen, it could be designed to compensate a non-linear interaction phase mismatch. The parameter m is an integer, which stands for the QPM order. For

odd m , the highest efficiency is attained with 50% duty cycle. First-order QPM assumes $m = 1$ and gives the highest efficiency, while higher-order interactions generate an efficiency proportional to $(2/\pi m)^2$.

Since QPM by periodic structures can be engineered for phase matching a single process, basic solutions were suggested in the form of two sequential gratings, the first for SHG, and the second would then perform SFG on the output products of the first grating. Two major drawbacks are characteristic of this method; the first is that it creates a component that can be used in a single direction only, as the doubling process should be done first. This limits the use in configurations such as optical parametric oscillators (OPOs) and other intra-cavity uses. The second drawback is that the efficiency η is also proportional to L (under optimal focusing conditions), the interaction length, for a bulk configuration and L^2 for the waveguide case, and so by using this method the gratings should share the length of the crystal. A more sophisticated solution was proposed in the form of quasi-periodic structures, originated from the Fibonacci series [13], and then generalized for any two QPM processes [14].

A simple but somewhat less flexible solution can be found by exploiting higher orders of QPM [15]. Higher orders may be used to phase match multiple processes by a single-period structure. This method allows the use of the full length of the crystal for both interactions, and is completely symmetric so that the device can be used in a multiple-pass configuration. The apparent limitation of this method is its dependence on the optical properties of the material to satisfy the two phase conditions presented in (1a) and (1b).

We investigated this by using the 14.5- μm -period PPLN waveguide. The third-harmonic product was expected to be in the order of 100 nW for the fully phase-matched case, where the enhancement of the waveguide is multiplied for both SHG and SFG processes occurring simultaneously. Since the silicon detector that we used was also sensitive to the strong second harmonic produced by the crystal, a way to separate the beams had to be found. The end face of the PPLN crystal was cut in a 15° angle, to avoid parasitic resonant effects, which would also allow the red and green products to exit the waveguide at different angles due to the dispersion of the material. Moreover, a thin-film dichroic beam splitter and an absorption green bandpass filter were used to isolate the third-harmonic beam. Figure 1 shows the experimental setup, in which a weak third-harmonic signal was detected and isolated from the strong second harmonic.

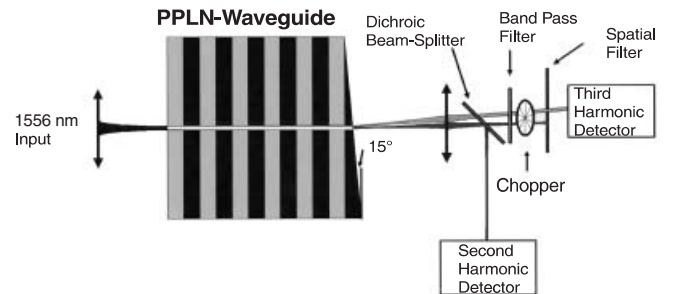


FIGURE 1 Experimental setup for separating the second- and third-harmonics generated in a PPLN waveguide

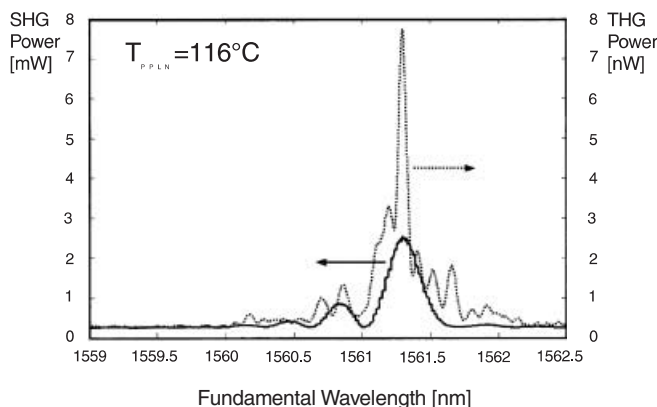


FIGURE 2 Second-harmonic (solid line) and third-harmonic (dotted line) generation in a PPLN crystal

The SHG and THG powers as a function of the fundamental wavelength are shown in Fig. 2, as they were measured at a crystal temperature of 87°C . Although a third-harmonic signal was detected, it showed lower than expected efficiency. A second Newport 778-nm external cavity tunable laser, co-linear with the 1550-nm fundamental wave, was coupled to the waveguide, to replace the second-harmonic beam and so the SFG process could be examined.

Results indicate that a non-phase-matched process was performed, and the efficiency function shows an irregular shape versus wavelength. The highest power level measured at the third harmonic was ~ 7.5 nW; therefore the SFG efficiency corresponds to an approximate interaction length of $300\ \mu\text{m}$ for a third-order QPM. Numeric calculations [16] suggest that the fundamental wavelength that was used in this experiment is about 100 nm longer than what would be required to fully phase match this interaction.

3 Spectroscopic measurements of $^{127}\text{I}_2$

3.1 Second-harmonic spectroscopy

The I_2 molecule's absorption lines near 775 nm are characterized by a low absorption coefficient, as the corresponding transitions originate in the 13 and 14 $X^1\Sigma_g^+$ vibrational bands. To populate the ground state, the absorption cell must be heated to about 800°K , while low pressure must be maintained to avoid pressure line broadening. By controlling a cold-point temperature of 40 – 60°C , the iodine pressure was several Torr. The absorption cell we used had quartz win-

dows, and did not show any sign of blackening, corrosion or any gas-purity problem.

Figure 3 shows the experimental setup used for SHG spectroscopy. First, a 760 – 790 -nm tunable diode laser was used, to test the cell's absorption properties, and to determine the conditions needed for sub-Doppler spectroscopy. The iodine was heated to 777 K, and the pressure was kept at 2.3 Torr. The waist $2\omega_0$ was $100\ \mu\text{m}$ and the highest power at the heated cell was 15 mW. The line observed is the $P(163)$ ($0 \leftarrow 13$) transition measured at $12856.7\ \text{cm}^{-1}$, limited by the 0.2-cm^{-1} accuracy of the wavemeter. Gerstenkorn et al. [9] measured this transition at $12856.6760\ \text{cm}^{-1}$, which is well within the error range of our measurement. The source was current-modulated at 20 kHz and demodulated by a lock-in amplifier to minimize the noise. The $3f$ derivative of the absorption signal was taken, and is presented in Fig. 4 where the sub-Doppler spectrum is easily identified. The lowest limit of saturation power that still allowed sub-Doppler observation was found to be 1.5 mW.

The 778 -nm laser was then substituted with a New Focus external cavity tunable laser, amplified by a 16 dBm EDFA and followed by a PPLN APE waveguide for SHG. The second-harmonic product was chopped at 1.5 kHz, and passed through the absorption cell. The lines were identified and compared to the atlas of [9], and the transitions were calculated and resolved according to published data [17], as can be seen in Table 1. Frequency scanning was done using the piezo-electric crystal of the laser to give the spectrum presented in Fig. 5. The envelope of periodic structure is probably the consequence of the etalon produced by the absorption cell end faces.

Rotational Transition	Vibrational Transition	Calculated Wavenumber [cm^{-1}]	Measured Wavenumber [cm^{-1}]	[9] Wavenumber [cm^{-1}]	[9] Line Number
R(28)	(0–14)	12849.379	12849.383	12849.371	4496
R(166)	(0–13)	12849.665	12849.655	12849.665	4497
R(27)	(0–14)	12849.698	12849.688	12849.703	4498
R(18)	(0–14)	12849.819	12849.820		
R(26)	(0–14)	12850.003	12850.002	12849.970	4499
R(17)	(0–14)	12850.118	12850.118		
R(25)	(0–14)	12850.294	12850.283		
R(16)	(0–14)	12850.404	12850.406		
R(15)	(0–14)	12850.677	12850.662	12850.653	4500

TABLE 1 Calculated and measured transitions of $^{127}\text{I}_2$ at 778 nm

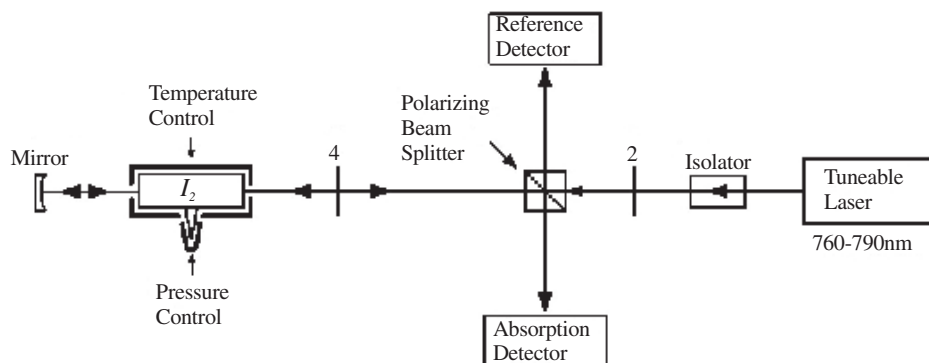


FIGURE 3 Experimental setup for sub-Doppler spectroscopy of iodine at 778 nm

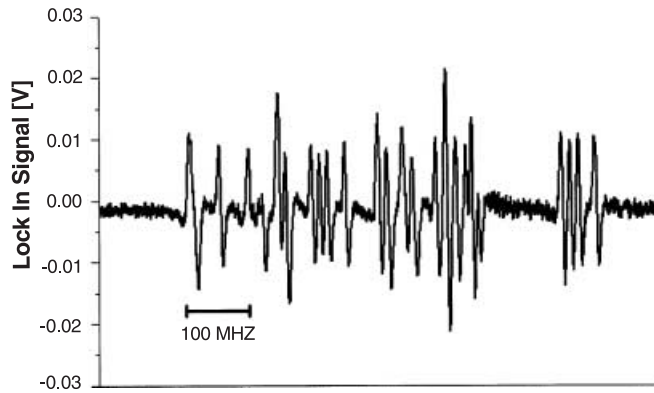


FIGURE 4 Third-derivative sub-doppler spectroscopy of iodine at 778 nm

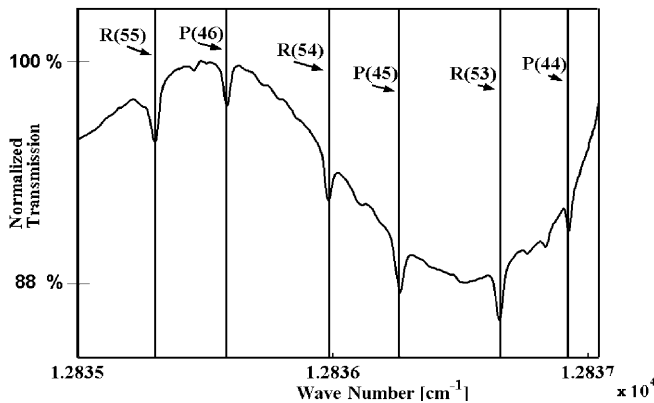


FIGURE 5 Spectroscopy by SHG of the $(0 \leftarrow 14)$ band of iodine near 778 nm. The vertical lines represent the corresponding calculated wavenumber for each one of the iodine transitions

These transition lines of the $(0 \leftarrow 14)$ band present low absorption coefficients. The measured value for the absorption coefficient of the $P(36)$ transition at $12\,842.1\text{ cm}^{-1}$ was found to have a value of $5.8 \times 10^{-5}\text{ cm}^{-1}\text{ Torr}^{-1}$.

We were not able to identify sub-Doppler lines with the 778-nm second-harmonic product, as the signal to noise levels were too low, and that remains for future work with more powerful pump lasers. Furthermore, anti-reflection coatings on the PPLN crystal, and angled windows for the absorption cell, may lower the noise levels and enable sub-Doppler stabilization of the 1556-nm laser.

3.2 Third-harmonic spectroscopy

Spectroscopic measurements of $^{127}\text{I}_2$ at room temperature were possible even with the weak third harmonic, as shown in a typical trace in Fig. 6. The strong absorption lines, originating in the vibrational transition between the $(40 \leftarrow 0)$ and the $(44 \leftarrow 0)$ bands, were compared to the literature [9] and found to be precise within the Burleigh wavemeter error range. Figure 6 also demonstrates the high density of transition lines, in which the average line spacing is about 5 GHz.

The absorption coefficient around 518 nm was measured in a cell held at room temperature, and the observed value was $0.51\text{ cm}^{-1}\text{ Torr}^{-1}$ for the $R(33)$ $(40 \leftarrow 0)$ line, about four orders of magnitude higher than the absorption measured in line $P(36)$ $(0 \leftarrow 14)$ at 778 nm.

To further understand the origin of this phenomenon, the relative population of the 778-nm transition ground state was

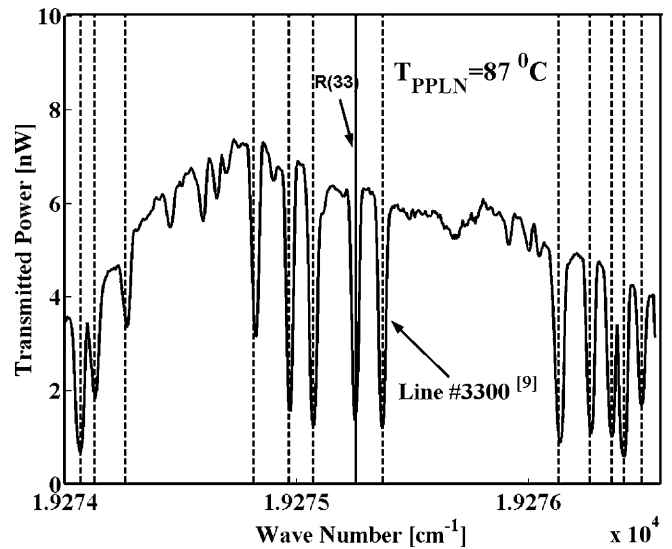


FIGURE 6 Spectroscopy by THG of iodine near 518 nm. The vertical lines represent the corresponding calculated wavenumber for each one of the iodine transitions. The absorption of the $R(33)$ $(40 \leftarrow 0)$ transition (solid line) was measured and found to be $0.51\text{ cm}^{-1}\text{ Torr}^{-1}$

estimated and found to be 2×10^{-5} smaller than that of the 518-nm transition; thus the higher absorption at the green is caused mainly by the difference in population of the ground state.

The absorption of iodine around 520 nm can be saturated by only a few hundred microwatts [18], so a stronger 1550-nm source could produce sufficient third harmonic for sub-Doppler spectroscopy.

4 Discussion and summary

This paper presents second- and third-harmonic spectroscopic measurements of iodine using a 1550-nm laser.

We have used a high-efficiency PPLN waveguide to produce a strong signal of 2 mW at 778 nm, which was used for spectroscopy of $^{127}\text{I}_2$, and the results are consistent with published calculations, taking into account the accuracy of the measurement equipment and that of the published articles.

Sub-Doppler spectroscopy of iodine was also performed by a 778-nm laser and third-derivative results are presented. Combining SHG and sub-Doppler techniques could significantly improve the number of available frequency standards for 1550-nm lasers, as iodine presents an average spacing of about 5 GHz between transition lines.

The second-harmonic product also served as a signal for the THG process which, although not phase-matched, was sufficient for third-harmonic spectroscopy of iodine at 518 nm. Basic calculations suggest that present 1400–1500-nm lasers could phase match this process, and based on currently existing high-power lasers at these wavelengths the system could provide enough third-harmonic power to achieve a sub-Doppler signal and further expand the frequency standards availability.

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