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# Molecular Iodine Spectra and Laser Stabilization by Frequency-Doubled 1534 nm Diode Laser

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By using the second harmonic radiation of a 1543 nm diode laser, several predicted rovibronic transitions of molecular iodine were observed for the first time. We demonstrated that the absolute frequency of a 1534 nm external-cavity diode laser could be frequency-stabilized to a 1 MHz instability (for one second averaging time) by locking the frequency of the second harmonic generation to the dense rovibronic spectrum of  $^{127}I_2$ . Our scheme shows one possible means of generating a frequency grid in the entire wavelength region of optical communications. [DOI: 10.1143/JJAP.44.3055]

KEYWORDS: 1550 nm, iodine molecule, DWDM, diode laser, optical communication

#### 1. Introduction

Frequency stabilized lasers with wavelengths in the region of optical fiber communications, are crucial in developing wavelength standard for use in dense wavelength-divisionmultiplexed (DWDM) systems. Stabilizing  $1.5 \,\mu$ m lasers referring to the transitions of various molecules and atoms, such as acetylene, hydrogen cyanide, rubidium, and potassium, has been performed and some promising results have been reported.<sup>1-6)</sup> However, it is not easy to find an appropriate reference molecule whose transitions have both a high density and a high resolution with the spectra covering the entire wavelength range of telecommunications, namely, from 1.5 to 1.6  $\mu$ m.

The rovibronic spectra of <sup>127</sup>I<sub>2</sub> provide rich frequency references within the wavelength range of the second harmonic generation (SHG) of the  $1.5\,\mu\text{m}$  to  $1.6\,\mu\text{m}$ radiations<sup>7–13</sup>) and are the most significant references in metrological applications.<sup>14</sup>) Thus detailed spectroscopic features of such spectra are extensively investigated in both theoretical and experimental. For example, around 800 nm, the calculations of all the absolute frequencies of <sup>127</sup>I<sub>2</sub> rovibronic transitions can be as precise as 100 kHz.<sup>7</sup>) Therefore, the absolute frequency references of DWDM systems can be realized whenever the corresponding laser is iodine-stabilized. The recently demonstrated comb laser in the optical-communication band<sup>15</sup>) shows great potential for experimentally determining the absolute frequency of iodine-stabilized DWDM systems.

In the past, probing iodine molecules by doubling the frequency of 1.5 to 1.6 µm radiations was difficult because the conversion efficiency of doublers was insufficient and the absorption of  ${}^{127}I_2$  in the 700 nm to 800 nm wavelength range was too week. Typically, a bulk periodically-poled LiNbO<sub>3</sub> (PPLN) crystal generates only few µW of second harmonic power under the condition of  $\sim 20 \,\mathrm{mW}$  fundamental power (~2%/W),16) however, at least few mW is needed for probing iodine transitions in the spectra of nearinfrared region.<sup>17)</sup> In 2000, owing to the development of a waveguide-PPLN,<sup>18,19)</sup> we reported on the observation of the rovibronic spectra of iodine molecules at around 767 nm by using the second harmonic generation of a 1.5 µm diode laser.<sup>20)</sup> In this study, we identified the above spectra and showed the promise of an "iodine reference" in DWDM systems by frequency-locking a 1534 nm diode laser to  $\sim$ 1 MHz instability within 1 second integration time. Our scheme demonstrated that a frequency-stabilized reference laser could possibly be fabricated with both a low frequency offset and a high resolution ( $\sim$ 1 MHz), which meet at fulfilling the wide-range requirements of DWDM systems. Moreover, green radiation, which as was generated from the waveguide-PPLN, was observed, and confirmed to be due to the self-sum frequency process<sup>17,21</sup>) by the presence of fundamental and second harmonic radiation in the waveguide-PPLN. Thus, using a single waveguide PPLN crystal, we showed the concept for the development of the absolute frequency references of three colors simultaneously.

## 2. Experimental Setup

The schematic diagram of our experiment is shown in Fig. 1, in which a tunable EOSI (Environmental Optical Sensors, Inc.) 2001 external-cavity diode laser of 2.7 mW output power was employed. Laser frequency could be tuned at 3.26 GHz/V by one piezoelectric transducer (PZT) and this PZT was dithered at 3.1 kHz to obtain the first-derivative signal of <sup>127</sup>I<sub>2</sub> spectra. Laser output power was coupled into an Erbium-doped fiber amplifier (EDFA) to coherently amplify the input radiation to a power level of 100 mW. To couple the output power of EDFA into the uncoated waveguide-PPLN efficiently, two lenses (L2, L3) of the same focal length (0.8 cm) were used, which resulted in a 60% coupling efficiency. The waveguide-PPLN, a 45mm  $\times$  $14 \text{ mm} \times 0.5 \text{ mm}$  crystal with a 10 µm mode field diameter, had several waveguide channels on the PPLN crystal with slightly different periods varying at the average value of 14.5 µm. A fiber polarization controller was employed to adjust the required input polarization, and a maximum second harmonic power of 8 mW was observed right after the output of PPLN crystal. At phase-matching temperature, the corresponding single pass conversion efficiency was 220%/W with a 0.27 nm full width half maximum (FWHM) bandwidth. Crystal temperature was controlled in the range from 60 to 120°C to change phase-matching wavelength and suppress the photo-refractive effect.<sup>22,23)</sup> To control crystal temperature under high temperature and high stability conditions, the temperature controller was designed with two stages: We implemented an on-off electronic heating system composed of a semiconductor relay for controlling the cool part of a TE cooler to  $95 \pm 0.1^{\circ}$ C, and an IXL Lightwave 5910B temperature controller for controlling the hot part to  $120 \pm 0.007^{\circ}$ C. Hence, by changing the temperature of the PPLN crystal, the corresponding phase-matching



Fig. 1. Experimental setup for iodine-stabilized diode laser system. Lenses used: L1: focal length f = 1.3 cm; L2, 3: f = 0.8 cm; L4: f = 5 cm; L5: f = 3 cm; L6: f = 40 cm; PBS: polarization beam splitter; PD1,2: photo diode; PSD: phase-sensitive detector.



Fig. 2. Phase-matched wavelength vs temperature of waveguide-PPLN. Note that the different channels of the waveguide yielded slightly different phase-matching wavelengths, and the highest discrepancy was 4 nm. That is, using the same crystal, the wavelength tuning range could be 11 nm by tuning the temperature of the frequency doubler.

wavelength could be adjusted from 1530 nm to 1537 nm, of the slope which is  $0.115 \text{ nm}/^{\circ}$ C, as shown in Fig. 2. The wavelength of the second harmonic radiation was measured using a Burleigh wavemeter with a resolving power of  $10^{6}$ . In our experiment, the tuning range of phase-matching wavelength was mainly limited by the optimum heating power of our temperature controller.

In Fig. 1, a chopper was installed before the iodine cell to further increase the signal-to-noise ratio (S/N) of the archived spectra. An iris was also installed to shape the beam profile and approximately 4 mW of round second harmonic radiation was obtained. The iodine cell was heated up to 650°C with a 25°C cold finger to increase the low-level population of the corresponding iodine transitions, thereby increasing the S/N of the spectra. The S/N did not significantly increase as a higher cell temperature was applied. The detectors (PD1, PD2 of silicon detectors) were confirmed that they do not respond to fundamental radiation. The aforementioned weak green radiation was also confirmed that it does not irradiate on PD2.

A residual power variation of SHG was observed as background among the archived first-derivative signals. This background was eliminated by balanced detection. That is, using the "subtracter" shown in Fig. 1 with a suitable gain, we could eliminate the slowly varying background by calibrating the output of the subtracter to zero in the absence of an iodine cell. The differential signal after the subtracter was demodulated at a laser dither frequency of 3.1 kHz by a phase-sensitive detector (PSD), and amplified by a lock-in amplifier at a chopping frequency of 410 Hz. To stabilize the laser, the error signal (output of lock-in amplifier) was fed back into the PZT of the diode laser through a laboratory-built proportional-integral (PI) feedback controller.

## 3. Results

Figure 3(a) reveals part of our resolved  $^{127}I_2$  rovibronic transitions at ~767 nm, which were archived from the first-harmonic output of the lock-in amplifier. The vertical axis is the strength of the rovibronic absorptions in arbitrary units while the horizontal axis is the controlling voltage of the PZT with a scan rate of 3.26 GHz/V. Figure 3(b) shows the corresponding first-derivative signal in which the optical modulation width is 130 MHz. Note that some small variations of the background in Fig. 3 are not noise but some unidentified weak transitions that appeared repeatedly. Table I shows the corresponding identified rovibronic transitions predicted by ref. 7, which are based on the

Table I. Corresponding rovibronic transitions in Fig. 3, referring to calculation results in ref. 7. ITU: International Telecommunication Union. The wavelength positions of those transitions are between two adjacent ITU grids, namely, 1534.25 nm and 1533.86 nm. Note that the average frequency spacing (<2 GHz) of those iodine transitions<sup>7)</sup> are one order of magnitude smaller than that of ITU grids (50 GHz).

Identification	measured in this work (nm)	predicted in ref. 7 (nm)	corresponding fundamental wavelength (nm)	frequency spacing (GHz)
R(48) 0-13	767.079	767.074	1534.16	-1.2737
R(245) 2-12	767.075	767.070	1534.15	-3.8213
P(39) 0-13	767.061	767.054	1534.12	$< 1  \mathrm{GHz}$
P(169) 0-12	767.058	767.051	1534.12	-3.8215
R(47) 0-13	767.044	767.038	1534.09	-1.2739
R(177) 0-12	767.041	767.037	1534.08	-2.5478
R(135) 1-13	767.028	767.024	1534.06	-1.2739
R(38) 0-13	767.023	767.019	1534.05	-1.2739
R(143) 1-13	767.018	767.014	1534.04	



Fig. 3. (a) Doppler-broadened absorption spectrum of  ${}^{127}I_2$  lines at approximately 767 nm (frequency doubling of 1534 nm):  $L_{cell} = 50 \text{ cm}; T_{cell} = 650^{\circ}\text{C}; T_{cold finger} = 25^{\circ}\text{C}$ . The vertical axis is the strength of the rovibronic absorptions in arbitrary units; The horizontal axis is the controlling PZT voltage at a scan rate of 3.26 GHz/V. (b) Corresponding first-derivative demodulation signals: dither frequency 3.1 kHz; modulation width of second harmonic radiation 131 MHz. Corresponding rovibronic transitions: see Table I.

previous experimental results.<sup>8–13)</sup> There were approximately 0.005 nm constant discrepancies between the identified and predicted values, which are suspected to have resulted from the instrumental offset of our wavemeter since this offset is independent of wavelength. Figure 3 and Table I together reveal feature of the dense spectra of molecular iodine, in which seven rovibronic transitions were resolved between two adjacent wavelength grids, namely, 1534.25 nm and 1533.86 nm of the International Telecommunication Union (ITU). Note that the average frequency spacing (<2 GHz) of those iodine transitions<sup>7)</sup> are one order of magnitude smaller than that of ITU grids (50 GHz). When the laser is frequency-locked to the center of the strong transitions shown in Fig. 3(b), the spectral signal compared with the fluctuation of the error signal (S/N) of the P(135) 1–13 transition, for instance, is  $\sim$ 600 for 1 s sampling time. That is, by analyzing the frequency fluctuation according to the S/N, an approximately 1 MHz instability of the fundamental frequency was obtained.<sup>24-27)</sup> In other words, under the assumption of white-noise limitation, the resolution of our frequency-stabilized 1534 nm laser system could be estimated using:  $\Delta f_{1534 \,\text{nm}} = (N/2S) \times \text{linewidth} =$  $0.5 \times (1/600) \times 800 < 1$  (MHz) at 1 second time constant, where the Doppler linewidth of each rovibronic transition is theoretically estimated as approximately 800 MHz.<sup>28)</sup>

Note that the accuracy of this laser system could not be estimated precisely unless the Allan variances of two similar laser systems are checked<sup>27</sup>) and inter comparisons are constantly performed.<sup>29</sup> Using our present experimental setup, we can estimate the accuracy of our experimental data

according to the spectral accuracy of our wavemeter, that is,  $\pm 0.002 \times 2 = \pm 0.004$  nm, which is sufficient for DWDM applications according to ITU-T G.692 grids.

A weak green radiation was observed in the second harmonic radiation. The power relation between the green radiation and the second harmonic generation (SHG) was confirmed to be linear, as shown in Fig. 4, in which wavelength was mismatched for 0.1 nm for the optimum output power of the green radiation. On the basis of Fig. 4, we suggest that the green radiation mainly resulted from the self-sum frequency effect in the waveguide-PPLN.<sup>17,21</sup>



Fig. 4. Self-sum-frequency generation in this experiment, in which wavelength was mismatched for 0.1 nm to the optimum output power of green radiation.

## 4. Discussion

We successfully demonstrated that a 1.5  $\mu$ m diode laser could be frequency-stabilized to 767 nm iodine lines, and thus is important for the applications of DWDM systems in telecommunications. Moreover, both the experimental results and the theoretical predictions<sup>7–13,17</sup> suggest that the similar strengths of the absorption spectra of molecular iodine can be resolved from 700 nm to 800 nm, and that the fundamental radiation can provide wide-band references in wavelength range of fiber communications, particularly in the region of the most important ITU-T anchor at #525, 1552.52 nm.<sup>6</sup>

There are many interesting topics for metrological applications: 1. By using Doppler-free saturation spectroscopy in the wavelength range of 770 nm, in which the natural linewidth is smaller than 5 MHz, one can expect to stabilize a diode laser referring to the hyperfine transitions of  ${}^{127}I_2$ with an uncertainty lower than 100 kHz.<sup>17)</sup> 2. In 2002, Cheng et al. successfully showed<sup>30)</sup> that, at green wavelengths, the molecule iodine has the dense hyperfine spectra with both high S/N and narrow linewidths (smaller than 100 kHz). Therefore, under an experimental setup similar to that used in this study but at a higher fundamental power, the "byproduct" of the output of the waveguide PPLN, namely, 511 nm green radiation, could be used to probe iodine hyperfine transitions as has been reported in reference 30; thus a diode laser could possibly be equipped with DWDM resolution even 3 orders of magnitude higher than that achieved in this study. Note that in this experiment, the period of PPLN (14.5 µm) was not designed for sumfrequency generation, the PPLN crystal was not AR-coated, and the pump power was only 100 mW. In other words, approximately 80 µW green radiation obtained using a 600 mW fundamental power is sufficient for probing hyperfine transitions of molecule iodine with a high S/N as demonstrated in ref. 26.

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