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

Wang-Yau Cheng, Jow-Tsong Shy
Department of Physics, National Tsing Hua University, Hsinchu 300, Taiwan, ROC

Tyson Lin, C.-C. Chou
Department of Photonics, Feng Chia University, Taichung 407, Taiwan, ROC

Abstract

By using the second harmonic radiation of an 1543-nm diode laser, several predicted rovibronic transitions of molecular iodine were observed for the first time. We demonstrated that the absolute frequency of an 1534 nm external-cavity diode laser could be frequency-stabilized to 1 MHz instability (1 second averaging time) by locking the frequency of second harmonic generation to the dense rovibronic spectrum of $^{127}$I$_2$. Our scheme shows a possible way on making the frequency grid in all the wavelength region of optical communication.
1. Introduction

Frequency stabilized lasers with their wavelength in the region of optical fiber communication, are crucial in developing the wavelength standards for using in the dense wavelength-division-multiplexed (DWDM) systems. Stabilizing 1.5 \( \mu \text{m} \) lasers to the transitions of various molecules and atoms, such as acetylene, hydrogen cyanide, rubidium, and potassium, had been performed and some promising results had been reported\(^1, 2, 3, 4, 5, 6\)\(^)\). However, it was not easy to find an appropriate molecule-reference which transitions were both dense and high resolution with the spectra covering the whole needed wavelength of telecommunication, namely, from 1.5 \( \mu \text{m} \) to 1.6 \( \mu \text{m} \).

As people know that the rovibronic spectra of \(^{127}\text{I}_2\) provides rich frequency references around the second harmonic generation (SHG) of 1.5 \( \mu \text{m} \) to 1.6 \( \mu \text{m} \) wavelength\(^7, 8, 9, 10, 11, 12, 13\)\(^)\) and the spectra of \(^{127}\text{I}_2\) are the most significant references in metrological applications\(^14\)\(^)\), that the detailed spectroscopic features are widely investigated both in theory and experiments. For example, around 800 nm, the calculations of the absolute frequencies of \(^{127}\text{I}_2\) rovibronic transitions can be as precise as 100 kHz.\(^7\)\(^)\) 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\(^15\)\(^)\) shows even more promising for experimentally determining the absolute frequency of iodine-stabilized...
In the past, probing iodine molecule by doubling the frequency of 1.5 µm to 1.6 µm radiations was difficult because the conversion efficiency of doublers were insufficient and the absorption of $^{127}$I$_2$ during 700 nm to 800 nm wavelength is too week. Typically, bulk PPLN (periodically-poled LiNbO$_3$) crystal generates only few µW of second harmonic power under ~ 20 mW fundamental power ($\sim$ 2%/W) $^{16}$ while at least few mW is needed for probing the iodine transitions in the near infrared region$^{17}$. In 2000, thanks to the development of waveguide-PPLN$^{18, 19}$, we reported on the observation of the rovibronic spectra of iodine molecule around 767 nm by the second harmonic generation of a 1.5 µm diode laser$^{20}$. In this paper, we identified those transitions and showed the promising of a “Iodine-reference” in DWDM by frequency-locking a 1534-nm diode laser to around 1 MHz instability under 1-second integration time. Our scheme demonstrated that a frequency-stabilized reference laser could possibly be made with both low frequency-offset and high resolution ($\sim$1 MHz), which fits the wide-range requirements of the DWDM system. Moreover, a green radiation was observed as was generated from the waveguide, and was confirmed as due to the self-sum frequency process $^{17, 21}$ by the presence of fundamental and second harmonic radiation in the waveguide-PPLN. Thus, using a single waveguide PPLN crystal, we showed the ideology of developing 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. The laser frequency could be tuned at 3.26 GHz/V by one piezoelectric transducer (PZT) and the PZT was dithered at 3.1 kHz in order to obtain the first-derivative signal of $^{127}\text{I}_2$ spectra. The laser output was coupled into an Erbium-doped fiber amplifier (EDFA) for coherently amplifying the input radiation to a power level of 100 mW. To couple the output power of EDFA into the uncoated waveguide-PPLN efficiently, two lens (L2, L3) of the same focal length (0.8 cm) were used, which resulted in 60% coupling efficiency. The waveguide-PPLN, a 45 mm $\times$ 14 mm $\times$ 0.5 mm crystal with 10 $\mu$m mode field diameter, had several waveguide channels on the PPLN crystal with slightly different periods varying around 14.5 $\mu$m. A fiber polarization controller was employed to adjust the required input polarization, and maximum second harmonic power of 8 mW was observed right after the output of PPLN crystal. Under an phase-matching temperature, the corresponding single pass conversion efficiency was 220%/W with a 0.27 nm FWHM (full width half maximum) bandwidth. The crystal temperature was controlled to range from 60$^0\text{C}$ to 120$^0\text{C}$ for changing the phase-matching wavelength and for reducing the photo-refractive
effect\textsuperscript{22, 23}). To control the crystal temperature in both high temperature and low instability, the temperature controller was designed with two stages: We implemented an on-off electronic heating system comprised of a semiconductor relay to control the cool part of one TE cooler to $95^\circ C \pm 0.1^\circ C$, while the hot part of the TE cooler was temperature-controlled up to $120^\circ C \pm 0.007^\circ C$ by an IXL Lightwave 5910B temperature controller. Hence, by changing the temperature of PPLN crystal, the corresponding phase-matching wavelength could be adjusted from 1530 nm to 1537 nm, which slope is 0.115 nm/\(^\circ C\) as shown in Fig. 2. The wavelength of the second harmonic radiation was measured by a Burleigh wavemeter with an resolving power of \(10^6\). In our experiment, the tuning range of phase-matching wavelength was mainly limited by the optima heating power which our temperature controller could offer.

In Fig. 1, a chopper was installed before the iodine cell for further increasing the S/N (signal-to-noise ratio) of the archived spectra. The iris was for shaping the beam profile and about 4 mW of round-shape second harmonic radiation was obtained. The iodine cell was heated up to 650 \(^\circ C\) with 25 \(^\circ C\) cold finger temperature to increase the lower level population of the corresponding iodine transitions, consequently, to increase the S/N of spectra. The S/N was not significantly increased as higher cell temperature was applied. The detectors (PD1, PD2, silicon detectors) were checked as no response to the fundamental radiation. The aforementioned weak green radiation
was also checked as did not irradiate on the PD2 detector.

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

3. Results

Figure 3a reveals part of our resolved $^{127}$I$_2$ rovibronic transitions around 767 nm, which were archived from the first-harmonic output of lock-in amplifier. The vertical scale stands for the strength of the rovibronic absorptions in arbitrary unit while the horizontal scale was the controlling voltage of PZT with the scan rate of 3.26 GHz/V. Fig. 3b is the corresponding first-derivative signal in which the optical modulation width was 130 MHz. Note that some small variations of the background in Fig. 3 were not noise but some unidentified weak transitions, which appeared repeatedly.
Table 1 shows the corresponding identified rovibronic transitions predicted by reference 7 which based on the former experimental results\(^8,9,10,11,12,13\). There were around 0.005 nm constant discrepancies between identified and predicted values, which was suspected as arose from the instrumental offset of our wavemeter since this offset was independent of wavelength. Fig. 3 and Table 1 together reveal the feature of 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 International Telecommunication Union (ITU). Note that the average frequency spacing (< 2 GHz) of those iodine transitions\(^7\) are one order of magnitude smaller than that of ITU grids (50 GHz). When laser is frequency-locked to the center of the strong transitions in Fig. 3b, the spectral signal comparing with the fluctuation of error signal (S/N) of P(135) 1-13 transition, for instance, is ~ 600 for 1 second sampling time. That is, by analyzing the S/N corresponding frequency fluctuation, around one MHz instability at the fundamental frequency was obtained.\(^24,25,26,27\) On the other words, under the assumption of white noise limitation, the resolution of our frequency-stabilized 1534 nm laser system could be estimated as:

\[
\Delta f_{1534\text{nm}} = \frac{1}{2} \times \frac{N}{S} \times \text{linewidth} = 0.5 \times \frac{1}{600} \times 800 < 1(\text{MHz}) @ 1 \text{ second time constant}
\]

Where the Doppler linewidth of each rovibronic transition was theoretically estimated as around 800 MHz\(^28\)
Note that the accuracy of this laser system could not be estimated precisely here unless Allan Variance of two similar laser system was checked and inter-comparisons were constantly performed. At the present status of our experimental set up, we can estimate the accuracy of our experimental date according to the spectral accuracy of our wavemeter, that is, \( \pm 0.002 \times 2 = \pm 0.004 \) nm, which is sufficient for the DWDM applications according to the ITU-T G.692 grids.

A weak radiation of green color was found among the second harmonic radiation. The power relation between the green radiation and the second harmonic generation (SHG) was checked as linear as shown in Fig. 4, in which the wavelength was mismatched for 0.1 nm for optima the output power of the aforementioned green radiation. Basing on the result of Fig. 4, we suggest that the green radiation was mainly resulted from the self-sum frequency effect in the waveguide-PPLN.

### 4. Discussion

In summary, we successfully demonstrated a 1.5-\( \mu \)m diode laser could be frequency-stabilized to the 767-nm iodine lines, which yields the significance in the applications of DWDM system in telecommunication. Moreover, both the experimental results and the theoretical predictions suggested that the similar strength of absorption spectra of molecular iodine can be resolved from 700 nm to 800 nm, that the fundamental radiation will provides a wide-band references in the
region of fiber communication, especially in the region of the most important ITU-T anchor, namely, #525, 1552.52 nm.

There also exists many interesting topics for metrological applications: 1. By using the Doppler-free saturation spectroscopy where the natural linewidth is smaller than 5 MHz, one can stabilize diode laser to the hyperfine transitions of the $^{127}\text{I}_2$, and fewer than 100 kHz uncertainties around 770 nm of SHG could be expected. 2. In 2002, W.-Y. Cheng et al. had successfully shown that, among the green wavelengths, the iodine molecule could offer a dense hyperfine spectrum with both good S/N and narrow linewidth (smaller than 100 kHz). Therefore, under similar experimental set up as was performed in this paper with higher fundamental power, the “by-product” of the output of waveguide PPLN, namely, the 511 nm green radiation, could be used to probe iodine hyperfine transitions as what was done in reference 30, and thus the diode laser could possibly be equipped with even 3 order of magnitude higher DWDM resolution than that of what was done in this paper. Note that in this experiment, the period of PPLN (14.5 µm) was not designed for sum-frequency generation, the PPLN crystal was not AR coated, and the pump power was only 100 mW. On the other words, if people could obtain ~ 80 µW green radiation by using a 600 mW fundamental power, it is already sufficient for probing hyperfine transitions of iodine molecule with good S/N as demonstrated in reference
The authors are grateful to Professor Martin M. Fejer and Dr. M. H. Chou of Stanford University for providing the waveguide-PPLN device. The authors also wish to thank the supports of the National Science Council of ROC under contract no. NSC 88-2112-M-007-042 and no. NSC 93-2112-M-259-002.
Figure Captions

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

Fig. 2 The phase-matched wavelength vs. the temperature of the waveguide-PPLN. Note that different channel of waveguide yielded slightly different phase-matching wavelength, and the most discrepancies could be 4 nm. That is, using the same one crystal, the wavelength tuning range could be 11 nm by tuning the temperature of the frequency doubler.

Fig. 3 (a) Doppler-broadened absorption spectrum of $^{127}$I$_2$ lines around 767 nm (frequency doubling of 1534 nm): $L_{\text{cell}} = 50 \text{ cm}$; $T_{\text{cell}} = 650 \degree \text{C}$; $T_{\text{coldfinger}} = 25 \degree \text{C}$; The vertical scale stands for the strength of the rovibronic absorptions in arbitrary unit; The horizontal scale was subjected to the PZT voltage with 3.26 GHz/V. (b) The corresponding first-derivative demodulation signals: dither frequency: 3.1 kHz; modulation width of second harmonic radiation: 131 MHz. The corresponding rovibronic transitions: see Table 1.

Fig. 4 The self-sum-frequency generation in this experiment, in which the
wavelength was mismatched for 0.1 nm to optima the output power of the green radiation.
The corresponding rovibronic transitions of Fig. 3, which refers to the calculating results of reference 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 are one order of magnitude smaller than that of ITU grids (50 GHz).
Table 1

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Temperature of PPLN, °C

Phase Matching Wavelength, nm

slope: 0.115 nm / °C

Fig. 2
Fig. 4

slope: 0.044 µW/mW
Reference


7) In this paper, the software of “IodineSpec 4” from Toptica Photonics Inc. was used, which based on the experimental results of reference 7 to reference 12.


9) S. Gerstenkom, P. Luc, J. Verges, J. Chevillard, “ATLAS DU SPECTRE D’ABSORPTION DE LA MOLECULE D’IODE”, Laboratoire Aime Cotton, CNRS II, 91405 Orsay (France) 11000 cm⁻¹ – 14000 cm⁻¹ (1982).


13) S. Gerstenkom, P. Luc, J. Physique 46 (1985) 867.


