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Express Letter

Sub-Doppler Spectroscopy of Molecular Iodine at 531 nm Using a Frequency-Doubled α-Distributed Feedback Laser

Tong-Long HUANG[1,2], Wang-Yau CHENG[1], Yan-Rung LIN[1], Jow-Tsong SHY[1] and Hai-Pei LIU[2]

[1]*Department of Physics, National Tsing Hua University, Hsinchu, Taiwan 300, R.O.C.*
[2]*Institute of Optical Sciences, National Central University, Chung-Li, Taiwan 320, R.O.C.*

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We report, for the first time, the observation of the iodine hyperfine transitions at 531 nm using a frequency-doubled α-distributed feedback (α-DFB) laser. The moderate high power of the α-DFB laser allows us to generate the second harmonic light by a periodically-poled $LiNbO_3$ single-pass frequency doubler. We can stabilize this laser frequency to the hyperfine component a_{10} of R(94) 34-0 or R(70) 33-0 I_2 line, and the preliminary frequency stability was about 5×10^{-11}. This laser system is an attractive frequency standard at 531 nm due to its compact size, high reliability, and low cost.

KEYWORDS: α-DFB laser, frequency-doubling, sub-Doppler spectroscopy, frequency standards, hyperfine structure, Doppler-broadened lines

1. Introduction

Several visible lasers stabilized to the hyperfine components of the molecular iodine (I_2) have been adopted as frequency/wavelength standards.[1)] Recently, there are many interests in the study of the hyperfine structure of iodine using green laser systems.[2)] The iodine shows strong absorption in this wavelength region. The iodine-stabilized green lasers, for example, 543 nm He–Ne laser[3)] and frequency-doubled 1064 nm Nd:YAG laser,[4)] have better performances than the commonly used iodine-stabilized 633 nm He–Ne laser system.

In this letter, we report, for the first time to our knowledge, the observation of the saturated absorption spectroscopy of I_2 hyperfine components near the 531 nm using a novel tunable laser system. This system includes an 1062 nm angled-grating semiconductor laser, also knows as α-distributed feedback (α-DFB) laser, and a single-pass periodically-poled $LiNbO_3$ (PPLN) monolithic frequency doubler. The α-DFB laser operates in a single frequency regime with high coherence and has impressive beam qualities and moderately high power.[5)] The PPLN doubler provides second harmonic power large enough for saturation spectroscopy of I_2 in single-pass configuration. Our laser system has a wide tunable range near 531 nm. Several Doppler-free spectra of iodine ro-vibrational transitions in 531.1774–531.2445 nm wavelength range were observed and assigned. We have stabilized the laser frequency on the a_{10} hyperfine component of the R(94) 34-0 and R(70) 33-0 transitions and preliminary frequency locking results will be presented, too. This laser system is an attractive frequency/wavelength standard at 531 nm due to its compact size, high reliability, and low cost.

2. Experiments and Results

Our experimental setup is shown in Fig. 1. An SDL-6752-P1 1062 nm α-DFB laser was used as the light source. Its output frequency can be tuned over 400 GHz by changing the injection current and the operation temperature. The current and temperature tuning coefficients are +0.36 pm/mA (or −96 MHz/mA) and +90 pm/K (or −24 GHz/K), respectively. This laser emitted radiation in 1062.29–1062.60 nm with power larger than 500 mW at 20°C for current from 1.6 to 2.6 A. Its output has a nearly circular beam profile with about 6 mrad full width at half-maximum (FWHM) divergence angle and no collimating lens is used in our experiment.

An optical isolator (OFR IOT-5-YAG-HP) is used to prevent the optical feedback effect. After passing through the optical isolator, the infrared light was focused by a lens of focal length of 150 mm into a spot size of 35μm inside a 45 mm long and 0.5 mm thick dual-wavelength anti-reflection (AR)-coated PPLN crystal. The crystal has a domain period of 6.5 μm and its temperature was controlled to about 170°C for quasi-phase-matching (QPM) frequency doubling. We obtained a second harmonic power of about 3 mW at 500 mW fundamental pumping power by single-pass frequency doubling. The measured conversion efficiency is 1.25%/W which is about 10% of the predicted conversion efficiency for the

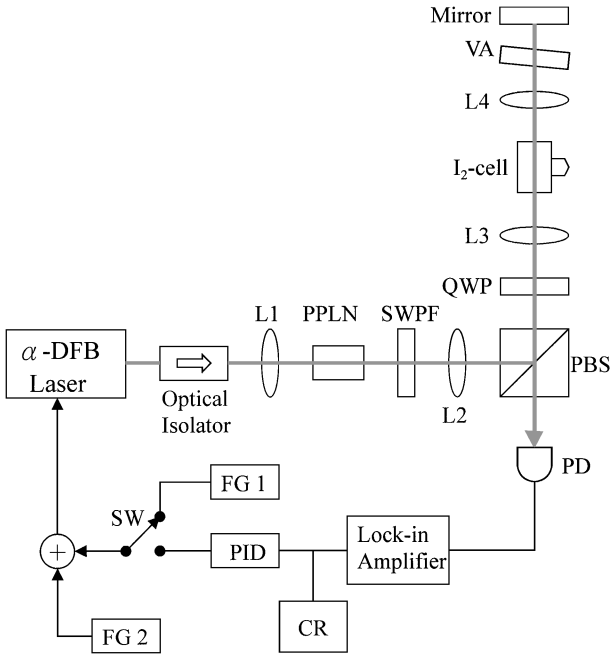

Fig. 1. Experimental setup for our 531 nm laser source and saturation spectroscopy of iodine. Here, PD: photodiode; PBS: polarizing beam splitter; QWP: quarter wave plate; VA: variable attenuator ; SWPF: short wavelength pass filter; L1: infrared lens with 150 mm focal length; L2, L3, and L4: visible lens with 100, 100, and 25.4 mm focal length, respectively; CR: chart recorder; FG 1 and FG 2: function generator; PID: proportional-integral-derivative loop; SW: switch.

perfect first-order QPM.[6] The poor efficiency is due to bad uniformity in domain periodicity. This means that we can obtain much higher second harmonic power using a better PPLN crystal. The green radiation can be generated in a frequency range over 800 GHz near 531 nm by controlling the α-DFB laser output wavelength and the PPLN temperature. Finally, the output of the PPLN crystal passes through a short wavelength pass filter (70% transmission) to eliminate the fundamental wave.

For obtaining the Doppler-broadened iodine spectrum, the second harmonic light was sent into a 6 cm long $^{127}I_2$ cell and detected by a photodiode. Figure 2 shows a 45 GHz (corresponding to 90 GHz at 531 nm) wide frequency scan of the iodine spectrum. The laser frequency was scanned by varying the injection current at 20°C operation temperature. In this case, we can observe fourteen relatively strong transitions, originating from the lowest vibrational level ($\upsilon = 0$) in the iodine X state. Table I list the line numbers, the measured and calculated frequencies, and the assignments of those transitions, as taken from the iodine atlas.[7]

The hyperfine structures of the Doppler-broadened lines were observed by using the sub-Doppler saturation spectroscopy and the third harmonic demodulation technique. After reflected by a polarizing beam splitter, the second harmonic light passes through a quarter-wave plate and the I_2 cell. The I_2 cell has a length of 6 cm and its cold finger is kept at 5°C using a thermoelectric cooler. The corresponding iodine vapor pressure in the cell is 7 Pa. Finally, a variable attenuator was used to decrease the strength of probe beam reflected from a mirror. The power densities of the pump and probe beams were about 15 mW/mm^2 and 1 mW/mm^2, respectively. The probe beam passed through the quarter wave plate and polarizing beam splitter and detected by an optical receiver. One function generator produced a slow triangle wave to vary the injection current for laser frequency tuning, the other a 7 kHz sine wave to modulate laser frequency. The hyperfine structure was obtained by a lock-in amplifier using the third harmonic demodulation and recorded by a chart recorder.

We have observed the hyperfine structures for all iodine lines in Table I, the number of observed hyperfine components (15 or 21) is in agreement with the predictions based on the selection rules for the rotational quantum number for transition from the X to the B state. In addition, we have calculated the hyperfine constants of each excited state using the Razet and Picard's empirical formula[8] and the hyperfine structures by the program provided by S. Picard.[9] The observed hyperfine structures agree well with the calculations.

In Fig. 3, a third derivative spectrum is shown for the hyperfine transition of line 1255 R(94) 34-0. The fifteen-hyperfine components, within the Doppler-broadened profile, are fully resolved. The calculated center frequency and the full span of the hyperfine structure are 18825.2021 cm^{-1} and 1064.9 MHz, respectively. Since the frequency drift at 531 nm is bout 100 MHz caused by the instability of the diode operation temperature. The frequency was scanned at a rate of 20 MHz/s. The recorded spectrum shows a "linewidth" of 20 MHz, wider than the true linewidth, due to the slow response of the chart recorder. The true linewidth should be under 2 MHz for most components.

Finally, the zero-crossing of the hyperfine component a_{10}

Table I. $^{127}I_2$ Absorption lines at 531 nm, within a tuning range of the frequency-doubled α-DFB laser. (Taken from ref. 7)

line	Measured (cm^{-1})	Calculated (cm^{-1})	Assignment
1248	18823.7283	18823.7256	P(68) 33-0
1249	18823.7949	18823.7959	R(27) 32-0
1250	18823.8289	18823.8301	P(24) 32-0
1251	18824.2969	18824.2964	R(137) 37-0
1252	18824.5637	18824.5645	R(26) 32-0
1253	18824.5986	18824.5969	P(23) 32-0
		18824.8660	P(123) 36-0
1254	18824.8877	18824.8884	P(109) 35-0
1255	18825.2031	18825.2021	R(94) 34-0
1256	18825.3049	18825.3027	R(25) 32-0
1257	18825.3306	18825.3335	P(22) 32-0
1258	18825.4973	18825.4961	R(70) 33-0
1259	18825.8730	18825.8689	P(67) 33-0
		18826.0110	R(24) 32-0
1260	18826.0396	18826.0403	P(21) 32-0
1261	18826.1038	18826.1016	P(91) 34-0

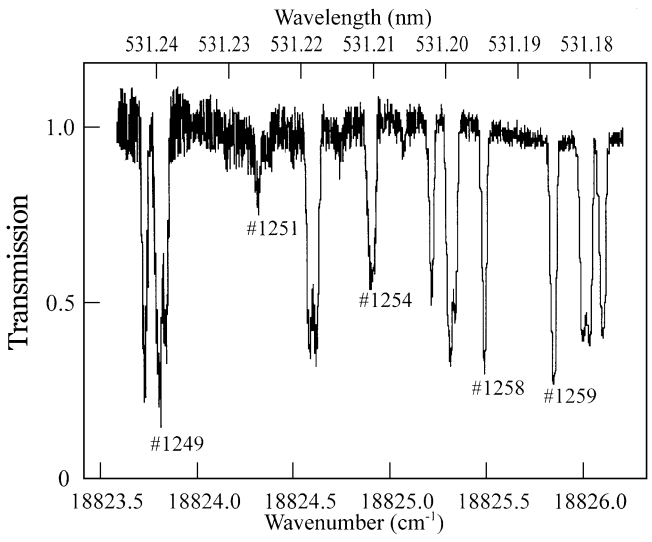

Fig. 2. Doppler-broadened lines of iodine in the wavelength range of 18823.5–18826.5 cm^{-1}.

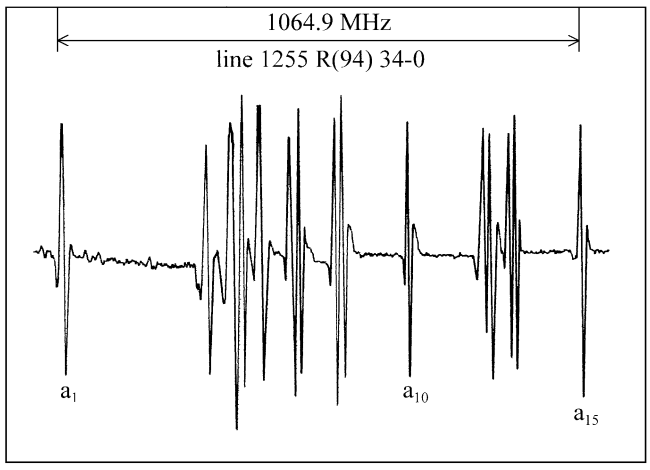

Fig. 3. Third derivative hyperfine spectrum of the R(94) 34-0 line in $^{127}I_2$.

of the more isolated lines R(94) 34-0 or R(70) 33-0 was used to lock our diode laser using a proportional-integral-derivative (PID) servo loop to control the laser current. The frequency stability was about 5×10^{-11} estimated from the error signal fluctuation after the laser was locked. We like to mention that the spectroscopic conditions, *e.g.*, iodine pressure, laser power density, and frequency modulation depth, and the PID loop were not optimized in this preliminary study.

3. Conclusions

We have observed the Doppler-free hyperfine components of molecular iodine at 531 nm with a compact tunable solid state laser source that includes an 1062 nm α-DFB semiconductor laser and a single-pass PPLN frequency doubler. The moderate high output power of the α-DFB allows us to directly generate the second harmonic light using a single-pass PPLN doubler, thus avoiding the complications due to enhancement cavity or power amplifier.[10] We have also locked its frequency on the iodine hyperfine component with a stability of 5×10^{-11}. The stability can be improved by optimizing the spectroscopic conditions and servo loop. Furthermore, the stability figure can be improved by the following: 1. Using extended-cavity configuration[11] to reduce frequency instability, 2. Using a PPLN having uniform domain period to increase the second harmonic power, and 3. Applying modulation transfer spectroscopy[12] or frequency modulation spectroscopy[13] to obtain frequency references having better signal to noise ratio. This laser allows one to build a compact frequency standard, potentially extendable to a wide wavelength range. We are going to use these I_2 lines to stabilize our laser for high precision IR spectroscopy of molecules using PPLN difference frequency generation (DFG) system.[10,14] Moreover, we will measure our stabilized laser frequency with respect to the iodine-stabilized frequency-doubled 1064 nm Nd:YAG laser in the future.

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