Japanese Journal of Applied Physics
Vol. 45, No. 4A, 2006, pp. 2776–2779

Measurement of Hyperfine Splitting of Molecular Iodine at 532 nm by Double-Passed Acousto Optic Modulator Frequency Shifter

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(Received February 18, 2005; revised December 9, 2005; accepted December 16, 2005; published online April 7, 2006)

The hyperfine splitting of molecular iodine R(56) 32-0 transition at 532 nm is measured using a double-passed acousto optic modulator frequency shifting apparatus. Preliminary results, compared with the values recommended by the Consultative Committee for Length, show that the accuracy of our method is better than 20 kHz, which can be easily improved in the future.
[DOI: 10.1143/JJAP.45.2776]

KEYWORDS: molecular iodine, acousto optic modulator frequency shifter, saturation spectrum, hyperfine structure, hyperfine splitting

1. Introduction

Molecular iodine (I_2) has been an attractive subject for spectroscopy because of its rich spectra from UV to IR. I_2 spectra have been shown to be suitable frequency references for laser frequency stabilization (see references in ref. 1). Moreover, I_2 also provides a good subject with which to test theoretical models of the Hamiltonian of hyperfine interactions.[2)] The measured hyperfine splitting is fitted to a four-term Hamiltonian and the corresponding hyperfine constants are obtained.

In general, hyperfine splitting is measured by the heterodyne technique. Two lasers are independently stabilized to different hyperfine components and their frequency difference is obtained by measuring beat frequency. However, not every laboratory could set up two iodine-stabilized lasers for measuring hyperfine splitting due to budget limitation or the commercial unavailability of such lasers. For example, we have observed the hyperfine structures of sixteen iodine transitions at 531 nm using a frequency-doubled 1062 nm alpha-distributed feedback diode laser.[3)] Because the manufacture of such an alpha-distributed feedback diode laser was discontinued by the manufacturer, we were not able to set up a second laser and measured hyperfine splitting using the heterodyne technique.

To solve this problem, we use a double-passed acousto optic modulator (AOM) frequency shifter to measure the hyperfine splitting of iodine. The frequency shifting property of an AOM has been widely used in laser frequency stabilization[2)] and laser spectroscopy.[4)] In our approach, laser frequency is stabilized to one hyperfine transition and the saturation spectrum of another hyperfine transition is measured using a double-passed AOM frequency shifter. In this paper, we present our preliminary test on this method by measuring the hyperfine splitting of the R(56) 32-0 transition of $^{127}I_2$. The hyperfine splitting of iodine lines near 532 nm has been measured precisely by the heterodyne technique using two iodine-stabilized lasers.[5,6)] The differences between our preliminary measurements and those recommended by Consultative Committee for Length (CCL) are less than 20 kHz. This indicates that our method is simple and useful for measuring hyperfine splitting using only one stabilized laser.

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2. Experimental

The schematic of our experimental setup is shown in Fig. 1. The experimental setup consists of three parts: laser frequency stabilization, AOM frequency shifter, and saturation spectroscopy for iodine.

We use a 450 mW nonplanar ring oscillator (NPRO) Nd:YAG laser (Lightwave Electronics Model 126) as light source. The NPRO laser has the following good characteristics: a narrow linewidth (less than 5 kHz), a low frequency drift (less than 50 MHz/h), and a wide single-mode tuning range (larger than 35 GHz). Its frequency can be tuned by changing the temperature of the laser crystal (thermal tuning) or applying a voltage on the piezoelectric transducer (PZT) attached to the laser crystal. The former has a long response time of a few seconds and the latter has a response bandwidth of 100 kHz.

Laser frequency is modulated with a width of 1.5 MHz by applying a 30 kHz sinusoidal wave to its PZT. The NPRO laser beam passes through a 30 dB Faraday isolator to prevent the optical feedback effect and is then focused onto a periodically poled $LiNbO_3$ (PPLN) crystal. A half wave plate ($\lambda/2$) is used to adjust light polarization to the z-direction of the PPLN crystal. The PPLN crystal is 50-mm long and 0.5-mm-thick with a 6.5 μm period. It is heated to 53 °C for quasi-phase-matched frequency doubling. The 532 nm second-harmonic power is higher than 8 mW. We divide the 532 nm second-harmonic light into two beams using a 1 : 4 beam splitter (BS).

We use the weaker laser beam to lock the laser to one hyperfine component of R(56) 32-0 transition. This laser beam enters a 10-cm long iodine cell as the pump beam after passing a polarizing beam splitter (PBS) and a quarter-wave plate ($\lambda/4$). The pump beam is collimated to 1.3 mm in diameter at the center of the iodine cell. The cold finger of the iodine cell is kept at 0 °C (corresponding to an iodine vapor pressure of 4.1 Pa). A fraction of the pump beam reflected by a partial reflection mirror (PRM) is detected by a photodiode (PD). The Doppler-free saturation spectrum of the hyperfine components of R(56) 32-0 transition is observed by the conventional third-harmonic demodulation method using a lock-in amplifier. The time constant of the lock-in amplifier is 30 ms. To stabilize the Nd:YAG laser, the third-harmonic output of the lock-in amplifier is fed into the thermal tuning via a servo control loop. The

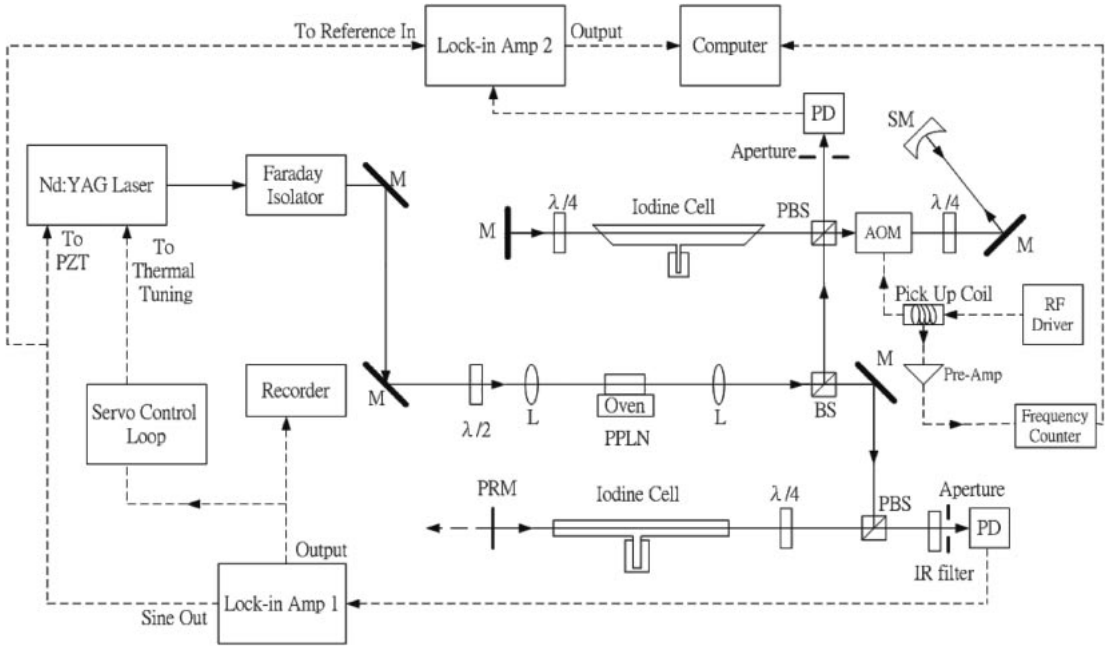

Fig. 1. Experimental setup for measuring hyperfine splitting of molecular iodine using double-passed acousto-optic modulator frequency shifter. M, mirror; $\lambda/2$, half-wave plate; L, lens; PPLN, periodically poled $LiNbO_3$; BS, beam splitter; PBS, polarizing beam splitter; $\lambda/4$, quarter-wave plate; PRM, partial reflection mirror; PD, photodiode; AOM, acousto optic modulator; RF, radio frequency; Pre-Amp, preamplifier; SM, spherical mirror (concave).

frequency stability of the Nd:YAG laser is better than 20 kHz.

The detailed working principle of the double-passed frequency shifter has been described in ref. 4. The stronger laser beam enters an AOM to generate a frequency tunable beam for the saturation spectroscopy of the hyperfine components of R(56) 32-0 transition. The frequency of the light diffracted by the AOM is changed by pf_{AOM} where p (positive or negative integer) is the diffraction order and f_{AOM} is the AOM carrier frequency. A variable-frequency RF driver allows the carrier frequency to be varied from 57 to 103 MHz. A frequency counter (HP 5316B) monitors the output of the RF driver and measures carrier frequency through a pick-up coil. Carrier frequency can be adjusted by transmitting a voltage to the RF driver and setting the tuning coefficient to about 3 MHz/V.

When the 532 nm laser beam first passes through the AOM, one of the diffracted beams is selected. Beyond the AOM, a quarter-wave plate ($\lambda/4$) converts the diffracted beam from the linear polarization state to the circular polarization state. Next, the spherical mirror retro reflects the diffracted beam passing through both the $\lambda/4$ plate and the AOM. The second pass through the $\lambda/4$ plate yields a linearly polarized beam with a polarization plane orthogonal to that of the input beam, allowing the PBS to separate the input beam from the double-diffracted beam. The input beam doubly passes through the AOM and is finally formed with the double-diffracted beam. We call such a system a double-passed AOM frequency shifter. Note that the diffracted angle of a diffracted beam changes with carrier frequency, a problem not seen in the double-passed AOM frequency shifter. The optical beam entering the double-passed AOM frequency shifter finally returns to the original optical path when going out from the AOM frequency shifter. To optimize AOM diffraction efficiency for the double-diffracted beam, the beam curvature at the spherical mirror is matched to the mirror curvature radius of 60 cm. For the first-order diffracted beam, the conversion efficiency is between 15 and 23%, and the frequency shift is between 114 and 206 MHz. For the second-order diffracted beam, the conversion efficiency is less than 5% and the twice-diffracted frequency shift is between 228 and 412 MHz.

Finally, we use the 532 nm laser beam from the double-passed AOM frequency shifter to observe the saturation spectrum of a hyperfine structure. The beam enters a 10-cm-long iodine cell with Brewster windows. It is about 2 mm in diameter at the center of the cell. Its cold finger is also kept at 0 °C. An arrangement similar to that used in the frequency stabilization of the Nd:YAG laser is employed. The third-derivative signal of the saturation spectrum is obtained using a lock-in amplifier with a time constant of 30 ms by adjusting the carrier frequency of the RF driver. Consequently, we can determine the hyperfine splitting.

Data acquisition is controlled by a LabVIEW program. AOM carrier frequency is scanned by changing the input voltage of its RF driver through a function generator (HP 33120A) under the control of the LabVIEW program. The frequency step of AOM carrier frequency is about 0.6 kHz, corresponding to 1.2 kHz for the first-order diffracted beam and 2.4 kHz for the second-order diffracted beam. For each data point, the outputs of the frequency counter and lock-in amplifier are taken after a waiting time of 2 s. Therefore, the frequency scanning speed is 0.6 kHz/s for the first-order diffracted beam and 1.2 kHz/s for the second-order diffracted beam, and it takes a few hours to complete a scan of 10 MHz.

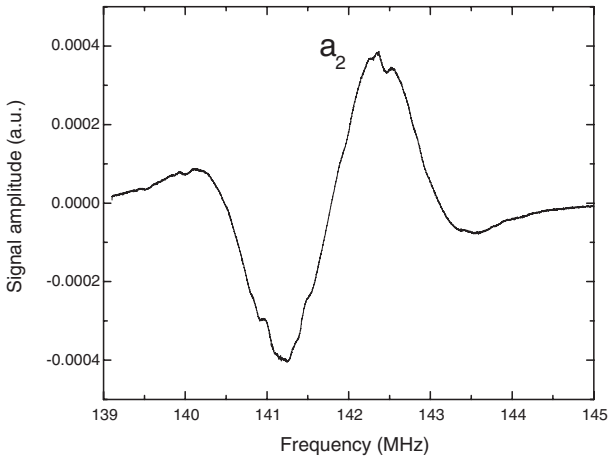

Fig. 2. Third-derivative saturation spectrum of a_2 component of R(56) 32-0 transition. We lock the Nd:YAG laser to the a_6 component and scan the a_2 component using the acousto optic modulator frequency shifter. The measured frequency interval between the a_6 and a_2 components is 141.789 MHz.

3. Results and Discussion

First, we lock the Nd:YAG laser to the a_{10} component of R(56) 32-0 transition and scan the a_6–a_9 and a_{11}–a_{14} components using the AOM frequency shifter. For the a_2, a_3, a_4, a_5, and a_{15} components, we lock the Nd:YAG laser to one component and scan the component within the tuning range of the first-order diffracted beam. Figure 2 shows the result in the case in which we lock the Nd:YAG laser to the a_6 component and scan the a_2 component. The frequency interval between the a_6 and a_2 components is 141 789 kHz, as determined by linear fitting to the portion near the center of a_2 component. The frequency interval between the a_1 and a_2 components is about 260 MHz; thus, we lock the Nd:YAG laser to the a_2 component and scan the a_1 component using the −2nd-order diffracted beam of the AOM.

The measured hyperfine splittings are listed in Table I. Using the a_{10} component as reference, the hyperfine splittings of R(56) 32-0 transition determined from our measurements are listed in Table II. The differences in hyperfine splitting between our results and CCL (1.5 kHz uncertainty) are also presented in Tables I and II. The discrepancy is below 20 kHz. The main contribution to the discrepancy is the frequency stability since we make only one measurement for each splitting (except for the a_{10}–a_{11} components, see below) listed in Table I. The frequency stability estimated from the error signal after the laser is locked is less than 20 kHz. Other contributions include pressure shift and impurities in different iodine cells, and different experimental conditions for frequency locking and saturation spectroscopy. To check experimental precision and accuracy, we lock the Nd:YAG laser to the a_{10} component and scan the a_{11} component fifteen times. The fifteen measured hyperfine splittings are averaged to 126.508 MHz with a standard deviation of 5 kHz. This value is comparable to the CCL value (126.513 MHz). The difference is only 5 kHz.

Generally, there is an offset between two lasers locked to the same hyperfine component due to differences in cells and operation conditions, and the offset in servo electronics. To eliminate this offset in hyperfine splitting measurement, in the conventional heterodyne technique, hyperfine splitting is determined first by measuring the beat frequencies of all hyperfine components with respect to a second stabilized laser and then obtained by simple subtraction. Since the span of hyperfine components is about 1 GHz, we are not able to measure the hyperfine splitting of all components with respect to a selected component. For example, the hyperfine splitting of the a_1 component to the a_{10} component is obtained by adding the results of three different measurements, which therefore result in a larger uncertainty. Besides, we are not able to remove the offset mentioned above. There are two other disadvantages of our method over the heterodyne technique: a larger laser power is required; diffracted beam power changes with AOM carrier frequency. The first may limit the application of our method and the second can be solved by intensity stabilization.

We will improve the accuracy of our method in the near future. For example, we can lock the voltage-controlled oscillator of the RF driver to the measured hyperfine peak and measure the hyperfine frequency more carefully. To

Table I. Measured hyperfine splittings of R(56) 32-0 transition.

Hyperfine splitting	Measured (MHz)	CCL (MHz)	Meas.–CCL (kHz)
a_1–a_2	259.701	259.698	3
a_2–a_6	141.789	141.780	9
a_3, a_4–a_7	131.128	—	—
a_5–a_9	143.990	143.977	13
a_6–a_{10}	170.066	170.064	2
a_7–a_{10}	154.547	154.548	−1
a_8–a_{10}	131.930	131.916	14
a_9–a_{10}	116.202	116.199	3
a_{10}–a_{11}	126.510	126.513	−3
a_{10}–a_{12}	131.223	131.212	11
a_{10}–a_{13}	154.503	154.488	15
a_{10}–a_{14}	160.682	160.665	17
a_{14}–a_{15}	125.747	125.747	0

Table II. Measured hyperfine splittings of R(56) 32-0 transition using a_{10} component as reference.

Hyperfine component	Measured (MHz)	CCL (MHz)	Meas.–CCL (kHz)
a_1	−571.556	−571.542	−14
a_2	−311.855	−311.844	−11
a_3, a_4	−285.675	—	—
a_5	−260.192	−260.176	−16
a_6	−170.066	−170.064	−2
a_7	−154.547	−154.548	+1
a_8	−131.930	−131.916	−14
a_9	−116.202	−116.199	−3
a_{10}	0	0	0
a_{11}	126.510	126.513	−3
a_{12}	131.223	131.212	11
a_{13}	154.503	154.488	15
a_{14}	160.682	160.665	17
a_{15}	286.429	286.412	17

reduce the error from the pressure shift and the impurities in different cells, we have fabricated a special cell in which two cells are connected by a tube. We will also try to reduce the offset of the servo electronics through better circuit design.

4. Conclusions

We have measured the hyperfine splitting of the R(56) 32-0 transition of molecular iodine at 532 nm using a double-passed AOM frequency shifter. Preliminary results, compared with the values recommended by the Consultative Committee for Length, show an accuracy better than 20 kHz. In the future, we will improve this method in terms of accuracy and use it to measure the hyperfine splitting of iodine lines at 531 nm, as observed using our frequency-doubled alpha-distributed feedback laser.[3)]

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