Frequency measurements and hyperfine structure of the $R(85)33-0$ transition of molecular iodine with a femtosecond optical comb

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Absolute frequency measurements of the $R(85)33-0$ transition of molecular iodine at the blue end of the tuning range of a frequency-doubled Nd:YAG laser are implemented with a femtosecond optical comb based on a mode-locked Ti:sapphire laser. The hyperfine structure of the $R(85)33-0$ transition is observed by use of high-resolution laser spectroscopy and is measured by the femtosecond optical comb. The observed hyperfine transitions are good frequency references for both frequency-doubled Nd:YAG and Nd:YVO_4 lasers in the 532-nm region. High-accuracy hyperfine constants are obtained by our fitting the measured hyperfine splittings to a four-term Hamiltonian, which includes the electric quadrupole, spin–rotation, tensor spin–spin, and scalar spin–spin interactions. © 2004 Optical Society of America

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1. INTRODUCTION

Diode-laser-pumped Nd:YAG lasers have been recognized as promising sources for high-resolution spectroscopy owing to their inherently low frequency noise, high reliability, and potentially high power. Nd:YAG lasers frequency stabilized on the hyperfine transition of molecular iodine are becoming important standards of optical frequency and wavelength. The absolute frequency of the $R(56)32-0{:}a_{10}$ hyperfine transition has been measured by a direct link between microwave and optical frequencies with a femtosecond (fs) optical comb.[1] A series of international frequency comparisons of iodine-stabilized Nd:YAG lasers has been carried out among several laboratories.[2–5] The 2001 meeting of the Consultative Committee for Length[6] led to a recommended value for the optical frequency of the $R(56)32-0{:}a_{10}$ hyperfine transition with an uncertainty of 5 kHz (relatively 9×10^{-12}) based on the reported frequency measurements.[1–3,7]

The hyperfine spectrum of iodine is extremely important for metrological applications. Many recommended optical frequency standards are based on the hyperfine transitions of molecular iodine.[6] Hyperfine structures of molecular iodine near 532 nm have been studied by several groups with improving accuracy.[8–13] The absolute frequency of 17 rovibrational transitions have been measured by use of an optical frequency comb generator, with the $R(56)32-0{:}a_{10}$ transition as a frequency reference.[14] These transitions are good candidates to be recommended for the practical realization of the meter.

An optical frequency comb generator usually has a frequency span of several terahertz,[15] which is efficient to bridge the frequency of iodine transitions near 532 nm but not enough for absolute frequency measurements. Photonic crystal fiber[16–18] (PCF) in combination with mode-locked fs lasers have revolutionized the research of broad-spectrum frequency comb generation and opened doors of many important applications including precision optical frequency metrology.[1,19,20]

The frequency stability of iodine-stabilized Nd:YAG lasers at 532 nm has reached a level of $<5 \times 10^{-14}$ at 1 s.[8,21] From the viewpoint of molecular physics, the high frequency stability and consequently attained high accuracy of the measurements have reduced the deviation of the theoretical fit of hyperfine splittings and hence increased the accuracy of the hyperfine constants. With accurate measurements of various hyperfine constants, we were able to derive or improve the formulas for hyperfine constants. One excellent example is the derivation of the rotation dependence of the ground-state (the X state)[22] and the excited state (the B state)[11] electric quadrupole hyperfine constants eQq'' and eQq', respectively. Recently we have also determined the vibration dependence of the tensor spin–spin and scalar spin–spin hyperfine constants by observing several relatively weak transitions.[12,13]

In the present paper we report, for the first time to our knowledge, the frequency measurement of the hyperfine structure of the $R(85)33-0$ transition at the blue end of the tuning range of the frequency-doubled Nd:YAG laser. Absolute frequency and hyperfine splittings of the transition are measured by use of a fs optical comb based on a mode-locked Ti:sapphire laser. Since the frequency of the observed transition is also close to the center of the tuning range of the frequency-doubled Nd:YVO_4 laser, the observed hyperfine transitions are important optical frequency references for both frequency-doubled Nd:YAG and Nd:YVO_4 lasers in the 532-nm region. A fit of the

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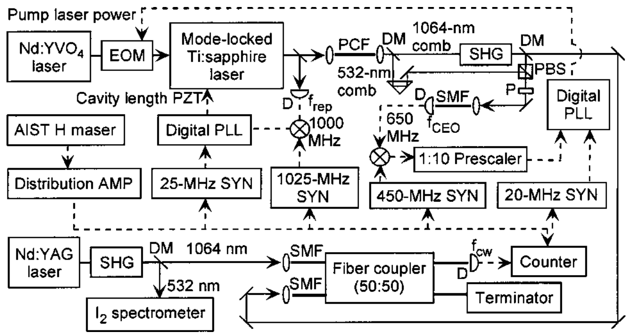

Fig. 1. Schematic diagram of the frequency measurement of an iodine-stabilized Nd:YAG laser that uses a fs optical comb. EOM, an electro-optic modulator; PCF, a photonic crystal fiber; DM, a dichroic mirror; SHG, second-harmonic generation; PBS, a polarization beam splitter; PZT, a piezoelectric transducer; PLL, a phase-lock loop; AMP, an amplifier; SYN, a synthesizer; SMF, a single-mode fiber; D, a detector; AIST, National Institute of Advanced Industrial Science and Technology.

measured hyperfine splittings to a four-term Hamiltonian is applied to the observed transition. High-accuracy hyperfine constants of the $R(85)33-0$ transition are obtained from this fit. The obtained hyperfine constants are important for further improving the precision of the formulas for hyperfine constants of molecular iodine.

2. EXPERIMENTAL SETUP

The experimental setup is shown in Fig. 1. A single-frequency Nd:YVO$_4$ laser with a maximum output power of 10 W (Coherent, Model Verdi-V10) was used as the pump source of a mode-locked Ti:sapphire laser (GigaOptics, Model GigaJet-30). The repetition rate and the width of the laser pulse were 1 GHz and 25 fs, respectively. An electro-optic modulator with a polarization beam splitter attached on the output side was inserted in between the pump laser and the mode-locked laser for the purpose of power modulation. A part of the output of the mode-locked laser was picked up by a photodetector for measuring the repetition rate ($f_{\rm rep}$). $f_{\rm rep}$ was beat down to approximately 25 MHz by use of a synthesizer with a fixed frequency of 1025 MHz. The beat-down frequency was fed to a digital phase-lock loop to control the length of the piezoelectric transducer attached on the mode-locked laser. All the synthesizers and counters used in the present experiment were phase locked to a H maser through a distribution amplifier.

Most of the output of the mode-locked laser was steered and focused into a commercially available PCF (Crystal Fibre, Denmark, Model NL-1.7-690). A f-to-$2f$ interferometer[20] was introduced to observe the carrier-envelop offset frequency ($f_{\rm CEO}$) of the fs comb. The green part of the broadened comb (532-nm comb) was separated out by a dichroic mirror and arranged to overlap with the second-harmonic generation of the infrared comb (1064-nm comb) at a polarization beam splitter. A delay line was introduced in the path of the 532-nm comb for the purpose of matching the timing of the two combs (which are actually fs pulses at the time domain). A KTiOPO$_4$ crystal of 7 mm was used for the second-harmonic generation of the portion of the comb near 1064 nm. A polarizer was used after the polarization beam splitter to adjust the power ratio of the two combs in the beat measurement. To ensure spatial-mode matching between the two combs, we introduced a single-mode fiber before the detector. The beat frequency, which is $f_{\rm CEO}$, was adjusted to a frequency around 650 MHz and mixed with a 450-MHz synthesizer to produce a frequency around 200 MHz. A 1:10 prescaler was introduced before a second phase-lock loop, which was used to servo control the $f_{\rm CEO}$ by tuning the pump power of the mode-locked laser through the electro-optic modulator.[23]

A diode-pumped Nd:YAG laser with a monolithic ring cavity (InnoLight, Model Prometheus-20NE) was used as a light source for iodine spectroscopy. The second-harmonic generation of the laser was obtained with a single-pass periodically poled KTiOPO$_4$ crystal. The spectroscopy of molecular iodine was based on the sub-Doppler technique of modulation transfer.[24,25] Frequency stabilization of the Nd:YAG laser based on the observed hyperfine transitions was realized. More detailed descriptions of the setup of our I$_2$ spectrometer and frequency stabilization are given elsewhere.[10–13,26]

The IR beam of the iodine-stabilized Nd:YAG was coupled into a single-mode fiber of a 50:50 fiber coupler for frequency measurement. The 1064-nm comb was also coupled into another port of the fiber coupler. The beat note between the cw IR light and the 1064-nm comb was observed at one output port of the coupler. Another output port was terminated to avoid interference due to the backreflected light from the fiber surface. The beat frequency was measured at 1064 nm by use of an avalanche photodetector followed by a frequency counter. All the measured frequencies are converted into green light frequencies in this paper.

3. EXPERIMENTAL RESULTS

A. Frequency Stabilization of the Femtosecond Optical Comb

Figure 2 shows the spectrum of the comb that was broadened by the PCF and was monitored by an optical spectrum analyzer. A frequency span of more than one octave has been achieved with the observed broadened comb. A non-polarization-maintaining PCF with a core diameter of 1.8 μm was used. The length of the PCF was

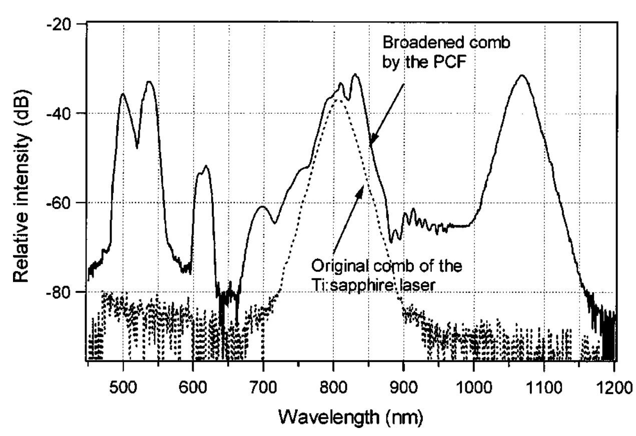

Fig. 2. Spectra of the original comb of the Ti:sapphire laser and the broadened comb by use of a PCF. The resolution bandwidth was 5 nm.

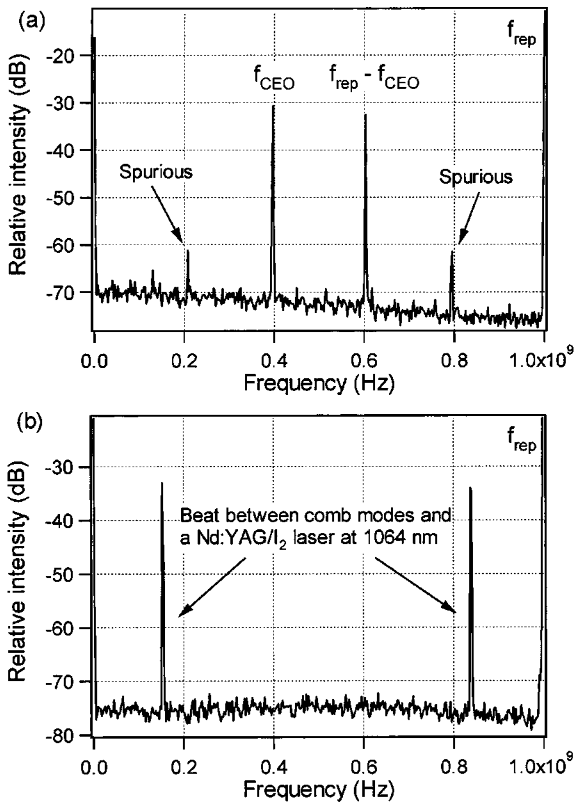

Fig. 3. (a) Observed CEO beat frequency. The resolution bandwidth was 300 kHz. (b) Beat frequency observed between an iodine-stabilized Nd:YAG laser and the broadened comb after the PCF at 1064 nm. The resolution bandwidth was 300 kHz.

approximately 50 cm. The average power of the original comb injected into the PCF was approximately 400 mW. The averaged power of the broadened comb observed after the PCF was approximately 120 mW. In the observed spectrum of the broadened comb, strong peaks were observed at 1064, 800, 532, and 500 nm, whereas holes were observed around 650, 580, and 950 nm. Although we cannot use the present comb to measure the optical frequencies at the wavelength regions in which the spectral holes exist, the strong peaks at 1064 and 532 nm enable us to observe both the f_{CEO} beat and the beat between the cw laser and the comb at 1064 nm. One important factor that affects the spectral structure of the broadened comb is the zero-dispersion wavelength of the PCF. The deep hole around 650 nm in the spectrum matches the zero-dispersion wavelength of the PCF. The pulse height of the present mode-locked laser with a repetition rate of 1 GHz is much lower compared with that of low-repetition-rate (~100 MHz) lasers. This is also one reason that causes the nonuniform spectra. The spectrum of the original comb of the Ti:sapphire laser is also indicated in Fig. 2 as a dashed curve.

Figure 3(a) shows the observed f_{CEO} and $(f_{\mathrm{rep}} - f_{\mathrm{CEO}})$ beat signals with the f-to-$2f$ interferometer. The signal-to-noise ratio (S/N) of the observed beat signals has exceeded 40 dB at a resolution bandwidth of 300 kHz and was sufficient for the digital phase-lock loop. In the present experiment the $(f_{\mathrm{rep}} - f_{\mathrm{CEO}})$ beat signal around 650 MHz was filtered out by use of a tunable filter and was used for frequency stabilization of the f_{CEO}. Once the f_{rep} and the f_{CEO} are phase locked, the whole frequency comb is phase locked to the H maser.

For evaluation of the frequency stability of the phase-locked optical comb, a beat measurement was implemented between the H-maser-locked comb and the iodine-stabilized Nd:YAG laser. Figure 3(b) shows the observed beat note between the comb modes and the cw laser light at 1064 nm. Again, the S/N of the observed beat signals has exceeded 40 dB at a resolution bandwidth of 300 kHz and was large enough to avoid miscounting in the frequency counter. Figure 4 shows the square root of the Allan variance[27] of the measured frequencies. The solid curve with triangles is for the measured beat frequency between the comb based on a H maser and the Nd:YAG laser locked on the $R(56)32-0{:}a_{10}$ transition. The a_{10} component of the $R(56)32-0$ transition was recommended by the Consultative Committee for Length for the practical realization of the definition of the meter and is an important target in the research of iodine-stabilized Nd:YAG lasers. We have established four iodine-stabilized Nd:YAG lasers including one transportable laser for investigations of the stability, repeatability, and reproducibility of Nd:YAG lasers based on the $R(56)32-0{:}a_{10}$ transition.[2,28–30] In Fig. 4 the dotted curve indicates the root Allan variance of our iodine-stabilized lasers, which was obtained by the beat measurement between two Nd:YAG lasers, both of which were locked on the $R(56)32-0{:}a_{10}$ transition. The typical stability of the H maser of the National Institute of Advanced Industrial Science and Technology is also indicated in Fig. 4 as a dashed curve. We notice that the Allan variance of the measured beat frequency between the H-maser-locked comb and the iodine-stabilized Nd:YAG laser basically follows the Allan variance of the H maser and that of the iodine-stabilized laser. For the short term (averaging time <20 s), the measured stability of the optical comb was basically limited by the H maser, although for the long term (averaging time >20 s), the

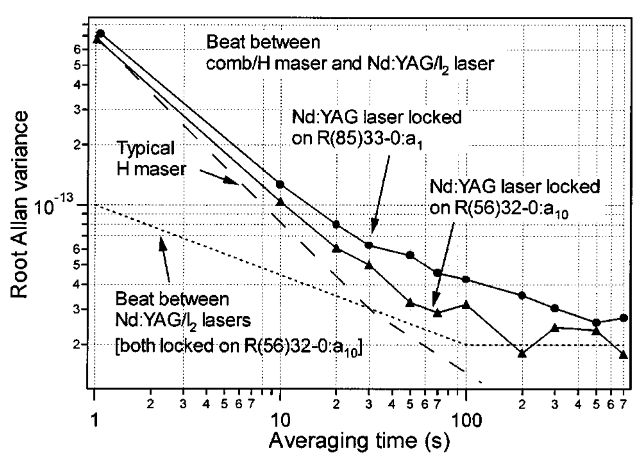

Fig. 4. Root Allan variance of the measured frequencies. The solid curve with triangles is for the measured beat frequency between the comb based on a H maser and the Nd:YAG laser locked on the $R(56)32-0{:}a_{10}$ transition. The solid curve with circles is for the measured beat frequency between the comb based on a H maser and the Nd:YAG laser locked on the $R(85)33-0{:}a_1$ transition. The dashed curve is for a H maser. The dotted curve is for beat frequency between two Nd:YAG lasers, both of which were locked on the $R(56)32-0{:}a_{10}$ transition.

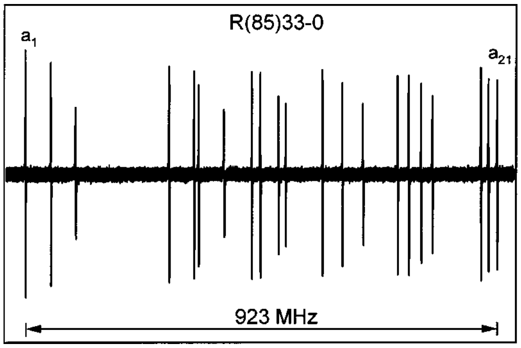

Fig. 5. Modulation transfer signals of the $R(85)33-0$ transition observed with a 1-kHz bandwidth.

measured stability of the optical comb was basically limited by the iodine-stabilized laser. Therefore, up to the measurement limit, we have confirmed that the optical fs comb is successfully phase locked to the H maser and can serve as an accurate frequency ruler for optical frequency metrology.

B. Observation and Frequency Measurements of Hyperfine Transitions

The frequency of the Nd:YAG laser depends on both the temperature of the laser crystal and the optical power of the pump diode. The laser frequency increases when we lower the temperature of the laser crystal or the power of the pump diode. By setting the temperature of the laser crystal at approximately 22 °C and reducing the laser power to 350 mW (32% of the full power), we can increase the laser frequency and access the $R(85)33-0$ transition at the blue end of the laser frequency tuning range. Figure 5 shows the observed hyperfine structure of the $R(85)33-0$ transition. The continuous frequency scan across the hyperfine structure was obtained by our tuning the temperature of the laser crystal slowly. In the modulation transfer spectroscopy the optical power of the pump and probe beams was 1.0 and 0.6 mW, respectively. Other experimental conditions are given elsewhere.[13] For odd J numbers of the ground rotational states, the rotational–vibrational energy level is split into 21 sublevels, resulting in 21 hyperfine components. In the observed $R(85)33-0$ transition, all the 21 hyperfine components are isolated from each other. The S/N of the a_1 component of the $R(85)33-0$ transition is approximately 115 in a 1-kHz bandwidth. The flat baseline of the observed spectra in the present experiment, which is significantly improved from previous observations,[31] is important for achieving high frequency stability and reproducibility.

The absolute frequency of each component of the $R(85)33-0$ transition was measured by use of the phase-locked optical comb. When the iodine-stabilized Nd:YAG laser beats with the nth comb mode at a frequency of f_{beat}, the absolute frequency of the Nd:YAG laser (f) can be calculated as

$$f = nf_{\text{rep}} + f_{\text{CEO}} \pm f_{\text{beat}}, \qquad (1)$$

where the f_{rep} and the f_{CEO} are phase locked to the H maser. In the actual experiment we must determine the integer n and consider two possibilities as shown in the above equation. Using a calibrated λ meter (Advantest, TQ8325), we could measure the absolute frequency of the Nd:YAG laser with an uncertainty of approximately 100 MHz, which is much smaller than the 1-GHz comb spacing. In this case, n is simply determined by solving Eq. (1) for n and requiring n to be an integer. The ambiguous sign of Eq. (1) is removed by one's observing the sign of the variation in the beat frequency f_{beat}, while one varies the f_{rep}.

Frequency stability of the measurement is an important fact that influences the measurement results. In Fig. 4 the solid curve with circles indicates the root Allan variance of the measured f_{beat} when the Nd:YAG laser was locked on the a_1 component of the $R(85)33-0$ transition. The measured frequency stability is approximately 1×10^{-13} for a 10-s averaging time, improving after a 300-s averaging time toward 3×10^{-14}. The long-term stability, which is limited by the iodine-stabilized laser, is slightly worse compared with that of the laser locked to the a_{10} component of the $R(56)32-0$ transition. This is because the $R(85)33-0$ transition is weaker compared with the $R(56)32-0$ transition, and, consequently, the S/N of the modulation transfer signal of the $R(85)33-0$ transition is smaller than that of the $R(56)32-0$ transition.

Figure 6 shows the result of ten measurements for the a_1 component of the $R(85)33-0$ transition during several days. The averaged frequency of the $R(85)33-0{:}a_1$ transition is 563 306 720 064.94 (6) kHz. Each measurement in the figure is an average of 30 to 100 beat frequency data points (depending on the measurement run), where each beat frequency data point was measured with a 10-s gate time by the frequency counter. The standard deviation of the beat frequency data for each measurement run was typically 70 Hz (relatively 1.2×10^{-13}). The frequency repeatability due to the process of locking and unlocking the laser is another important issue in the experiment. To check the repeatability, we have repeated ten measurements during several days (as shown in Fig. 6). The standard deviation of the ten measurements is approximately 60 Hz (relatively 1.1×10^{-13}), which indicates the repeatability of our measurement and is limited by the repeatability of the iodine-stabilized Nd:YAG laser. The accuracy of the H maser of the National Institute of

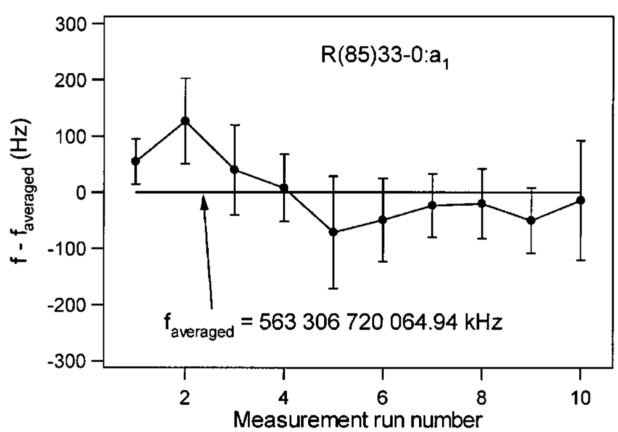

Fig. 6. Measured absolute frequency of the a_1 component of the $R(85)33-0$ transition. The uncertainties are given as one standard deviation of the averaging.

Table 1. Observed and Calculated Hyperfine Splittings of the $R(85)33\text{–}0$ Transition[a]

Hyperfine Component	Observed (kHz)	Calculated (kHz)	Observed Less Calculated (kHz)	Weight	F'	I
a_1	0	0	0	1.0	81	5
a_2	50732.5	50731.9	0.6	1.0	86	1
a_3	99742.3	99742.9	−0.6	1.0	91	5
a_4	281946.2	281947.1	−0.9	1.0	82	5
a_5	331678.7	331679.1	−0.4	1.0	85	3
a_6	341087.6	341087.4	0.2	1.0	87	3
a_7	389099.9	389099.8	0.1	1.0	90	5
a_8	445205.3	445206.3	−1.0	1.0	83	5
a_9	461608.4	461609.8	−1.4	1.0	84	3
a_{10}	496293.9	496294.2	−0.3	1.0	88	3
a_{11}	510619.4	510619.6	−0.2	1.0	89	3
a_{12}	582132.0	582132.2	−0.2	1.0	83	3
a_{13}	621988.5	621988.3	0.2	1.0	86	3
a_{14}	662825.5	662825.5	0.0	1.0	89	5
a_{15}	729463.3	729463.8	−0.5	1.0	84	5
a_{16}	751718.8	751718.1	0.7	1.0	85	5
a_{17}	777078.3	777077.3	1.0	1.0	87	5
a_{18}	798584.8	798584.0	0.8	1.0	88	5
a_{19}	892318.3	892318.4	−0.1	1.0	85	1
a_{20}	906642.5	906642.3	0.2	1.0	86	5
a_{21}	922692.5	922692.4	0.1	1.0	87	1

[a] The measurement uncertainty of the observed hyperfine splittings is 100 Hz. The standard deviation of the fit is 660 Hz.

Advanced Industrial Science and Technology was calibrated by the Coordinated Universal Time and confirmed to be better than 1×10^{-14} during the measurements.

For measurement of the hyperfine splittings, the Nd:YAG laser was locked in succession to all the 21 hyperfine components of the $R(85)33\text{–}0$ transition. An absolute frequency measurement was applied to each component. Table 1 shows the measured hyperfine splittings obtained by calculation of the frequency difference between each component and the a_1 component. Each observed result in the table is an average of more than 30 beat frequency data points, where each beat frequency data point was measured with a 10-s gate time by the frequency counter. As discussed previously, the repeatability of the iodine-stabilized laser, which ranges from 60 to 100 Hz depending on the S/N of the hyperfine transitions, has the largest contribution to the measurement uncertainty.

4. CALCULATION OF HYPERFINE CONSTANTS

We use the measured hyperfine spectra to determine the hyperfine constants for the observed transitions. The Hamiltonian of the hyperfine interactions, H_{hfs}, is written as[32,33]

$$H_{\text{HFS}} = eQq \cdot H_{\text{EQ}} + C \cdot H_{\text{SR}} + d \cdot H_{\text{TSS}} + \delta \cdot H_{\text{SSS}}, \tag{2}$$

where H_{EQ}, H_{SR}, H_{TSS}, and H_{SSS} represent, respectively, the electric quadrupole, spin–rotation, tensor spin–spin, and scalar spin–spin interactions and eQq, C, d, and δ represent the corresponding hyperfine constants for each of these interactions. We followed the procedure outlined by Bordé *et al.*[33] to calculate the eigenstates of the hyperfine Hamiltonian. In calculating the electric quadrupole interactions, we take ΔJ up to ± 4 into account. The rotational Hamiltonian H_R,

Table 2. Fitted Hyperfine Constants

Constant	$R(85)33\text{–}0$
ΔeQq (MHz)	1906.832(1)
ΔC (kHz)	94.9180(5)
Δd (kHz)	−48.78(8)
$\Delta \delta$ (kHz)	−10.19(6)

$$\langle JIF|H_R|JIF\rangle = BJ(J+1) - DJ^2(J+1)^2 + HJ^3(J+1)^3, \tag{3}$$

was introduced in this calculation, where I is the total nuclear spin and F is the total angular momentum. The necessary rotational constants B, D, and H are taken from the literature.[34] By theoretically fitting the observed spectra of the transitions, one can obtain accurate values for ΔeQq, ΔC, Δd, and $\Delta\delta$ but only a very crude estimate of the absolute values for the respective ground and excited states.

In the present calculation the hyperfine splittings are fitted to the measurements by use of a least-squares fit, where the hyperfine constants eQq', C', d', and δ' are varied. The lower-level constant eQq'' is derived from J dependence of eQq'',[22] and the lower-level constants C'', d'', and δ'' are fixed to the values given in the literature.[35]

The calculated hyperfine splittings and their differences from the observed values of the $R(85)33\text{–}0$ transition are listed in Table 1. The agreement between experiment and theory is excellent where the standard deviation of the fit is approximately 660 Hz, which is among the lowest standard deviations obtained for a theoretical fit. The fitted hyperfine constants for the $R(85)33-0$ transition are listed in Table 2. Hyperfine constants ΔeQq, ΔC, Δd, and $\Delta\delta$ with very low uncertainties are obtained from the theoretical fit.

Accurate hyperfine constants are important for establishing formulas that can be used to reproduce or simulate the hyperfine spectrum in iodine. In our previous paper[12] we derived *ab initio* formulas for the vibration dependence of the excited-state tensor spin–spin and scalar spin–spin constants d' and δ', respectively. In the fitting procedure, there were only two pairs of Δd and $\Delta\delta$ available for $v' = 33$, determined from the studies of the $R(84)33-0$ and $P(87)33-0$ transitions. The averaged value of Δd and $\Delta\delta$ for $v' = 33$ was $-48.74(29)$ kHz and $-10.32(38)$ kHz, respectively, where the quoted uncertainties are the differences between the constants obtained from the two transitions. By including also the constants of the $R(85)33-0$ transition (results of the present paper), we obtain for $v' = 33$:

$$\Delta d = -48.75(14) \text{ kHz}, \tag{4}$$

$$\Delta\delta = -10.28(20) \text{ kHz}, \tag{5}$$

where the quoted uncertainties are the standard deviation of the averaging of the constants obtained from the three transitions. The uncertainties of the constants have been reduced to approximately one half of the previous ones.

5. DISCUSSION

As determined by the 2001 meeting of the Consultative Committee for Length, the frequency uncertainty of the 532-nm iodine-stabilized Nd:YAG laser is 5 kHz.[6] Frequency differences can arise from differing saturation parameters, residual linewidths, and possible (small) contamination of the I_2 reference cells. The wave-front difference of the overlapping beams inside the iodine cell may also cause frequency shifts.[36] However, the above systematic frequency shifts can be neglected when we study the frequency difference between different transitions (f_{interval}) instead of the absolute frequency of each transition. The uncertainty of f_{interval} is much smaller compared with that of absolute frequency and is basically limited by the repeatability of the iodine-stabilized lasers, which is typically approximately 100 Hz depending on the S/N of the transition. For the $R(85)33-0$ transitions, f_{interval}, referring to the $R(56)32-0{:}a_{10}$ hyperfine transition, was obtained by measurement of the absolute frequency of both the $R(85)33-0$ and the $R(56)32-0$ transitions with the same laser under the same operation conditions (Table 3). From the viewpoint of metrological applications, the observed hyperfine transitions enrich the existent optical frequency reference network near 532 nm (a total of 20 rovibrational transitions),[13,14] especially at the blue end. The absolute optical frequency of each hyperfine transition (f) can be written as

$$f = 563\,260\,223\,513(5) \text{ kHz} + f_{\text{interval}} + f_{\text{HFS}}, \tag{6}$$

where 563 260 223 513(5) kHz is the absolute frequency of the $R(56)32-0{:}a_{10}$ component[6] and f_{HFS} is the measured hyperfine splitting. These references are excellent candidates for the practical realization of the meter.

Table 3. Measured Frequency Interval of the Observed Transition

Line	Assignment	f_{interval} (kHz)
1114	$R(85)33-0{:}a_1$	46 496 559.1
1110	$R(56)32-0{:}a_{10}$	0.0

Although the observed iodine transition is close to the blue end of the tuning range of a frequency-doubled Nd:YAG laser, it is close to the center of a frequency-doubled Nd:YVO_4 laser. The Nd:YVO_4 laser is especially attractive for establishing a compact and low-cost frequency-stabilized laser owing to its high pump efficiency. An iodine-stabilized Nd:YVO_4 laser is being developed in our laboratory for practical applications including length measurements. The observed transition in the present experiment should also serve as an optical frequency reference for the frequency-doubled Nd:YVO_4 laser.

Accurate frequency measurements are important for the determination of molecular constants. As shown in Section 4, hyperfine constants of molecular iodine are obtained for the $R(85)33-0$ transition. With the accumulating information, we can obtain a good understanding of the rotation and vibration characteristics of all the hyperfine constants. Furthermore, as being realized for hyperfine constants, the improved frequency stability, reproducibility, and measurement accuracy should also lead to improved rovibrational constants of molecular iodine. Research along this line is in progress.

The wavelength range in which we can use our fs optical comb for frequency measurements is now limited by the spectral holes observed in the broadened comb. Especially, the deep hole around 650 nm is close to the wavelength of the popular iodine-stabilized He–Ne laser at 633 nm. To improve the uniformity of the broadened comb, it is better to use a PCF with a zero-dispersion wavelength near the center of the original Ti:sapphire comb (800 nm). It is also helpful for obtaining a better uniformity in the comb to reduce the repetition rate of the mode-locked Ti:sapphire laser and, consequently, increase the peak power of each pulse. Since a usual commercially available λ meter can reach an accuracy of better than 100 MHz, it is reasonable to set the repetition rate of our laser somewhere in between 500 MHz and 1 GHz. Experiments along this line are also in progress.

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