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Absolute-Frequency Measurement of the Iodine-Based Length Standard at 514.67 nm

R. J. Jones
University of Colorado Boulder

Wang-Yau Cheng
National Dong Hwa University

K. W. Holman
University of Colorado Boulder

Lisheng Chen
University of Colorado Boulder

John L. Hall
University of Colorado Boulder, jhall@jila.Colorado.EDU

See next page for additional authors

Authors
R. J. Jones, Wang-Yau Cheng, K. W. Holman, Lisheng Chen, John L. Hall, and Jun Ye

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R.J. JONES
W.-Y. CHENG*
K.W. HOLMAN
L. CHEN
J.L. HALL
J. YE✉

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JILA, National Institute of Standards and Technology and University of Colorado, Boulder CO 80309-0440, USA

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ABSTRACT The absolute frequency of the length standard at 514.67 nm based on the molecular iodine ($^{127}I_2$) transition of the P(13) 43-0 component a_3 is measured using a self-referenced femtosecond optical comb. This frequency-based technique improves measurement precision more than 100 times compared with previous wavelength-based results. Power- and pressure-related frequency shifts have been carefully studied. The measured absolute frequency is 71.8 ± 1.5 kHz higher than the internationally accepted value of $582\,490\,603.37 \pm 0.15$ MHz, adopted by the Comité International des Poids et Mesures (CIPM) in 1997.

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1 Introduction

The definition of the unit of length and its practical realization are based on the adopted value for the speed of light, $c = 299\,792\,458$ m/s, and the frequency of an optical transition. Thus, length measurements are intrinsically related to the unit of time. Among the listed optical frequency standards recommended by the Comité International des Poids et Mesures (CIPM) in 1997 [1], molecular iodine (I_2) holds a unique position in that it offers five reference lines that have been most widely used for metrological calibration. Although the system at 514.67 nm has the promise of being one of the better-quality standards based on I_2, historically it has been probed only with cumbersome Ar^+ laser systems and the transition frequency was determined only through wavelength interferometry. The recommended 514.67-nm length standard was based on those measurements on the a_3 component of the transition P(13) 43-0 of $^{127}I_2$. The adopted value by the CIPM in 1997 is 582 490 603.37 MHz, with a standard uncertainty of 0.15 MHz (2.5×10^{-10}), for an iodine-cell cold-finger temperature set at −5 °C [1]. In this paper, we report the first absolute-frequency determination of this length standard, using a phase-coherent optical frequency comb linked to the Cs primary clock, the current realization of the unit of time. We report a measurement precision 100-times-improved over previous wavelength-based results.

Our motivation for performing detailed studies of hyperfine transitions of I_2 in the wavelength range of 500–532 nm is two-fold. The search for the best candidates of I_2-based optical frequency references requires systematic studies of the line width and strength of many transitions. Frequency-doubled Nd:YAG/$^{127}I_2$ at 532 nm has proved to be one of the most practical optical frequency standards due to its compact size, reliability, and high stability ($< 5 \times 10^{-14}$ at 1 s) [2]. In order to attain a higher frequency stability, it is useful to explore I_2 transitions at wavelengths below 532 nm, where the natural line widths may decrease at a faster rate than that for the line strengths as molecular iodine approaches its dissociation limit. Secondly, a widely tunable laser system permits systematic studies of rotation–vibration dynamics and hyperfine interactions near the dissociation limit, providing rich information on molecular structure and dynamics. Specifically, a large range of ro-vibrational quantum states can be accessed, allowing a detailed parametric study of transition strengths, hyperfine interactions, and collision physics. In work previously reported [3], we have built such a widely tunable, yet high-resolution and high-sensitivity I_2 spectrometer and measured the line widths of transitions within the range 523–498 nm. Signals were recovered with an excellent signal-to-noise ratio (S/N). We observed a clear trend of line-width narrowing with decreasing transition wavelength as molecular iodine approaches the dissociation limit. However, this tendency was complicated by variations in line widths among different rotational or hyperfine components. The limit on lifetime imposed by pre-dissociation and its associated broadening of the transition line width is being studied. We also discovered that the hyperfine patterns are dramatically influenced by the pre-dissociation effect [3]. Frequency-based measurements of these hyperfine intervals across a large spectrum will reveal important information on

✉ Fax: +1-303/492-5235, E-mail: ye@jila.colorado.edu
**Present address:* Department of Physics, National Dong Hwa University, Taiwan, Republic of China

the variations of charge distribution and molecular configuration. Our results indicate that I_2 transitions in the wavelength range 532–501 nm hold great promise for future development of optical frequency standards, especially when coupled with the all-solid-state Yb:YAG laser.

Since the introduction of Kerr-lens mode-locked femtosecond (fs) lasers for optical frequency metrology [4], wide-bandwidth optical combs have revolutionized the precision and procedure of optical frequency measurements, impacting an extensive range of optical frequency standards based on transitions of atoms, ions, or molecules [5–8]. Typically in a frequency measurement, the repetition rate, f_{rep} (or mode spacing of the comb), of the fs laser is phase-coherently locked to the Cs microwave-frequency standard. The other degree of freedom associated with the fs comb, i.e. the carrier–envelope offset frequency, f_{ceo}, can also be measured or stabilized with the same microwave standard using a self-referencing approach [6] or another known optical standard. This drastically simplified frequency-measurement scheme can be realized with a reliable and compact table-top system. The measurement accuracy of the fs optical comb system has been carefully studied [7, 8].

2 Iodine spectrometer

Figure 1 shows our experimental scheme that implements precision scan and control of the laser frequency based on a highly stable optical cavity augmented by the Doppler-free I_2 resonance recovered from the spectrometer. The fs comb system used for frequency measurement is also shown. Our single-mode Ti:sapphire laser can tune from 953 nm to 1080 nm, and at 1030 nm provides about 300 mW of useful output power. A small portion of the fundamental power is used for laser-frequency pre-stabilization to an evacuated, vibration-isolated, and thermally stabilized optical cavity. The operational laser line width is limited by the vibration noise associated with the cavity. Most of the Ti:sapphire laser power is used for second-harmonic generation (SHG) to probe I_2 transitions. The I_2 spectrometer is configured for frequency-modulation (FM) spectroscopy, with additional chopping of the pump beam to further reduce the influence of the Doppler background. The size of both pump and probe beams is 3 mm in diameter. We typically maintain $< 100\,\mu$W for the probe beam while the pump-beam power can be varied from 1 to 6 mW to study power-related shift and broadening of I_2 transitions. The high-purity I_2 cell was prepared by the Bureau International des Poids et Mesures (BIPM) and it has an 8-cm useful length with Brewster windows at both ends. Control of the I_2 pressure is maintained by the temperature stabilization of the cell's cold finger and is used for studies of pressure-related effects. For frequency measurement of the P(13) 43-0 a_3 transition, we use a cold-finger temperature of $-5\,°$C, in compliance with the CIPM recommendation.

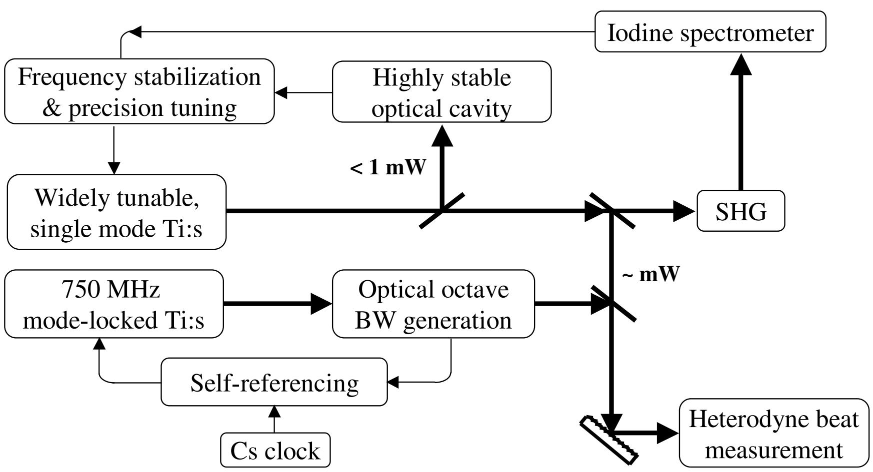

FIGURE 1 A precision I_2 spectrometer based on a widely tunable Ti:sapphire laser. The fs comb system used for frequency measurement is also shown

For the long-term laser-frequency stabilization via the I_2 resonance, an acousto-optic modulator (AOM) is placed between the laser and the pre-stabilization cavity. Drift and low-frequency noise on the laser stemming from the pre-stabilization cavity can be basically suppressed by feeding the error signals derived from the I_2 resonance into the drive frequency of the AOM. The laser light, which now carries the information of the I_2 resonance, is then directed to the frequency-measurement section based on a fs laser comb. Since there is more power available in the fundamental laser light near 1030 nm, it is used for frequency measurement.

3 Femtosecond optical-comb-based absolute-frequency measurement

The Kerr-lens mode-locked fs laser used for the optical frequency comb generation has a repetition rate of 750 MHz and produces an optical comb spectrum spanning ~ 30 nm, centered at 800 nm [9]. An octave bandwidth of the comb is generated by coupling 100 mW of laser light through a 20-cm-long micro-structure fiber [10]. Both f_{rep} and f_{ceo} are stabilized with reference to the Cs standard, thus establishing an absolute optical frequency grid of 750-MHz spacing, extending from 500 nm to 1100 nm. In practice, an intracavity piezo-activated mirror is used to control f_{rep} while the pump power for the fs laser is used to control f_{ceo}. The frequency of the mth comb component is given by $f_m = m f_{rep} + f_{ceo}$. The absolute frequency of the I_2-stabilized cw laser can thus be determined by counting its heterodyne beat frequency against a corresponding comb line. The comb order, m, is determined from a medium-resolution (500-MHz) wavelength meter.

To check the reliability of the 750-MHz fs comb, we use a frequency-doubled Nd:YAG laser stabilized on a $^{127}I_2$ transition (R(56) 32-0, a_{10}) at 532 nm as a calibration tool [2, 12]. The absolute frequency of the 532-nm system has been measured for over two years with a standard deviation less than 120 Hz at 1064 nm [13]. Previous measurements were carried out using a well-characterized 100-MHz fs

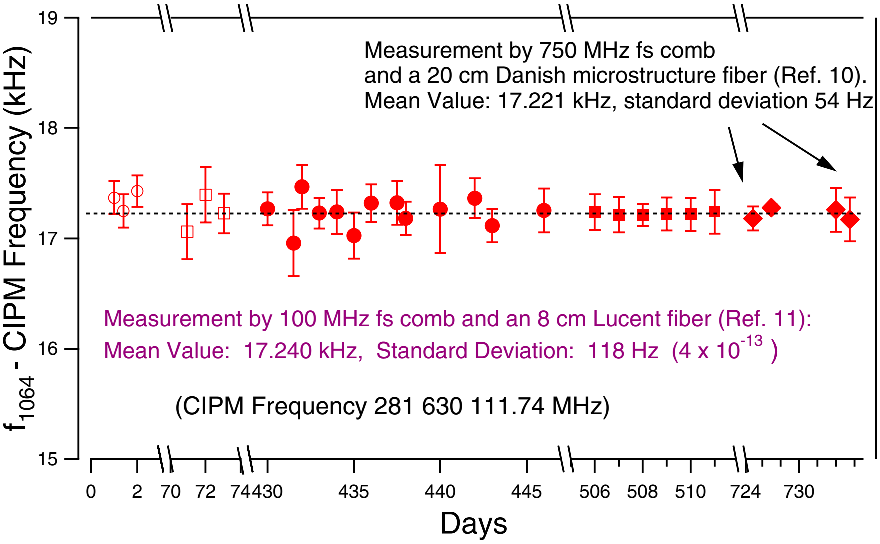

FIGURE 2 Augmented long-term frequency-measurement record of the Nd:YAG/I_2 532-nm system. Agreement with previous measurements based on 100-MHz systems with different micro-structure fibers [11] is excellent

laser system referenced to the Cs clock. Figure 2 shows the augmented long-term measurement record of the absolute frequency of R(56) 32-0 a_{10}. The last four data points represent measurements of the same I_2 transition using the 750-MHz comb system. The agreement between the two systems is within 20 Hz. At the present stage, the frequency-measurement noise of the 532-nm system is limited by the instability of the Cs clock (5×10^{-12} at 1 s). We are currently also working with a H-maser referenced optical comb system.

A remarkable feature of the present fs comb system is its reliability. It is now possible to acquire hours of measurement data without interruption. This will be important for the future development of an optical clock [13, 14]. Furthermore, the signal-to-noise ratio of the beat signal between the cw laser and the comb exceeds 45 dB within a 100-kHz bandwidth, permitting a direct frequency count without further signal processing. Figure 3b shows a frequency-measurement record of the length-standard transition, i.e. $^{127}I_2$, P(13) 43-0 a_3. The corresponding Allan deviation determined from the measurement record is presented in Fig. 3c. It is clear that the measurement noise is dominated by the instability of the Cs clock as well, just as that for the 532-nm system.

A preliminary estimate of the system performance at 514.67 nm can be made based on the S/N of the recovered resonance signal. Figure 3a shows the line shape of the $^{127}I_2$ P(13) 43-0 a_3 transition recovered using frequency-modulation spectroscopy with a modulation frequency of 6 MHz and a 5-ms time constant. The cold-finger temperature of the I_2 cell is near −5 °C, corresponding to 2.38 Pa vapor pressure [15]. The projected frequency noise of a laser locked to this error signal can be estimated from the discrimination slope and the rms noise at the baseline. The estimated frequency noise would be 3 kHz at 514.67 nm, corresponding to a fractional noise of 5×10^{-12} at 5 ms, or 3.5×10^{-13} at 1 s, with an 8-cm-long cell. Extending the I_2 cell length or placing the cell inside a multi-pass cavity will certainly improve the system performance.

The capability of absolute-frequency measurement greatly facilitates characterization of systematic effects of the spectrometer. While traditionally one employs a heterodyne beat technique between two similar systems to characterize optical standards, we can now calibrate the performance of a single system against the fundamental Cs standard. We have carefully studied frequency shifts depending upon the following experimental parameters: optical power, modulation frequency and amplitude, pressure, and optical alignment. To minimize subjective influence in the frequency measurement, we typically have a few experimenters align the spectrometer independently, with the only common objective being maximization of the signal size. Residual amplitude modulation (RAM) in FM spectroscopy is a usual source for systematic errors. We minimize the RAM on the frequency-modulated probe beam by careful adjustments of polarization optics throughout the spectrometer. When the RAM is minimized, we can em-

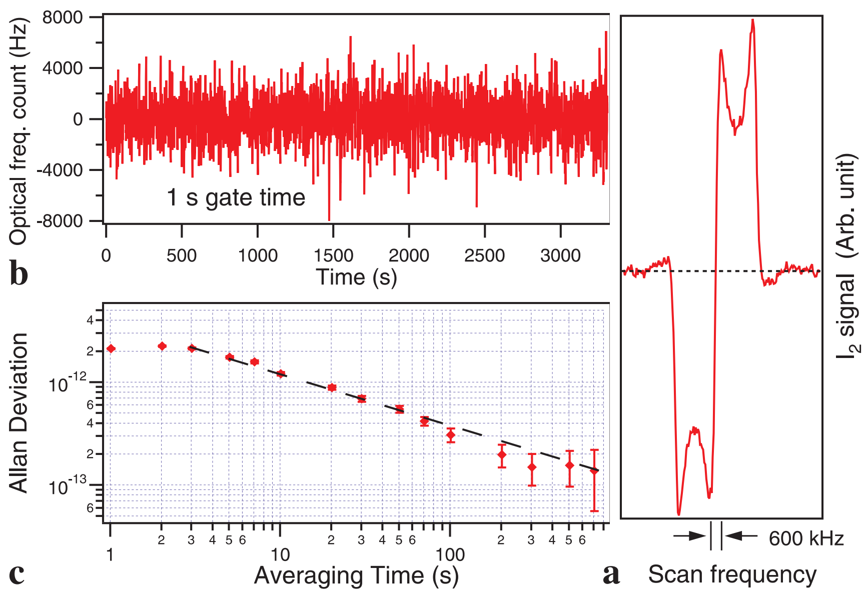

FIGURE 3 **a** Signal line shape recovered from FM spectroscopy of the transition P(13) 43-0 a_3. Cell cold-finger temperature near −5 °C, 5-ms averaging time. **b** Direct frequency measurement of P(13) 43-0 a_3 by the Cs-referenced fs comb. Gate time 1 s. Standard deviation is limited by the Cs clock. **c** Allan variance is determined from the time record in **(b)**

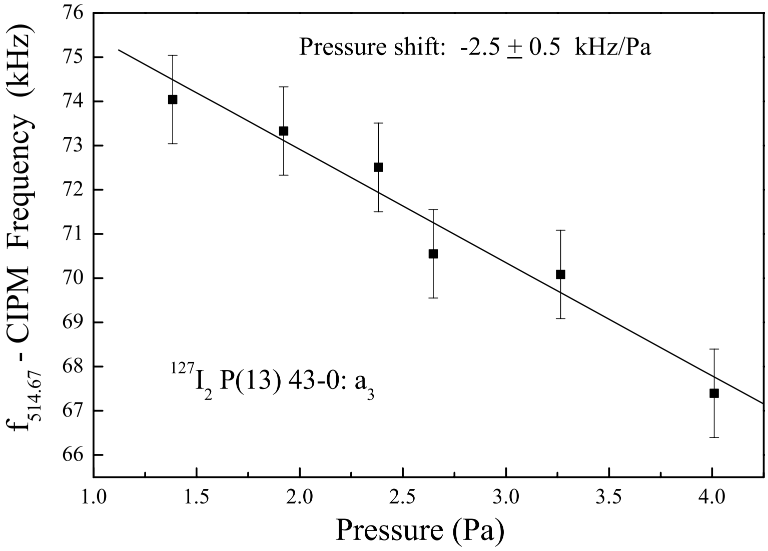

FIGURE 4 Pressure shift of the P(13) 43-0 component a_3

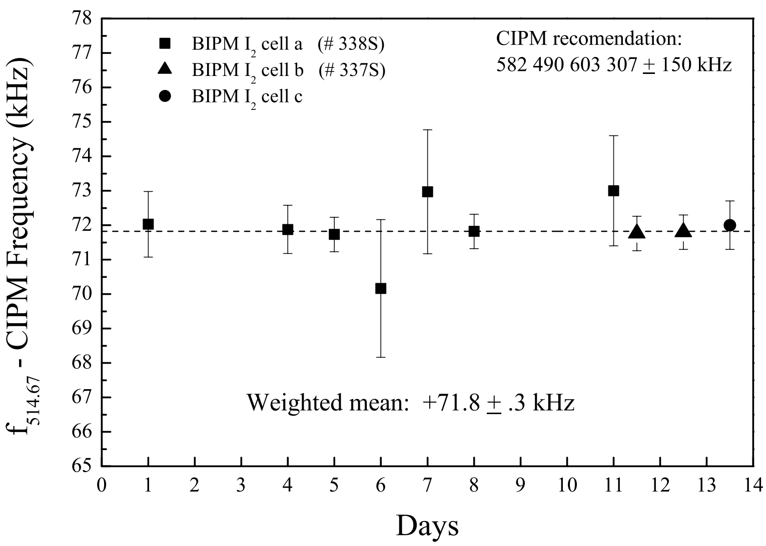

FIGURE 5 Long-term frequency-measurement record of the P(13) 43-0 component a_3. Three different I_2 cells are used in the measurement, as indicated by data points in *square*, *triangle*, and *circle*. The BIPM-designated number for cells a and b is shown in parenthesis

ploy different values of modulation frequency (4, 6, 8, and 10 MHz, for example) with various modulation amplitudes and still obtain consistent and reproducible frequency-measurement results. Within this optimized operation regime, optical-power-related frequency shifts are small, usually less than the measurement standard deviation, in the normal operation range of 1 mW to 4 mW for a 3-mm-diameter pump beam. By a controlled variation of the I_2 cold-finger temperature, we change the sample density in the cell. The related pressure-dependent frequency shift is measured for this length-standard transition, with a linear shift of -2.5 ± 0.5 kHz/Pa, as shown in Fig. 4.

Figure 5 shows a two-week frequency-measurement record of $^{127}I_2$ P(13) 43-0 a_3, with an average value of 71.8 ± 0.3 kHz higher than the CIPM-recommended value of $582\,490\,603\,370 \pm 150$ kHz [1]. To include other unforeseeable effects on the transition frequency, we quote a 5-sigma value of ± 1.5 kHz for the measurement uncertainty. To be more specific, further evaluations should include studies of beam size and wavefront curvature, signal-recovery techniques other than FM spectroscopy, and a large collection of various I_2 cells. However, we emphasize that even for the present work, we have used three different I_2 cells. Two of the cells were made in the same recent run while a third one was made more than eight years ago, also at BIPM. We observe no statistically significant difference among these three cells, as shown in Fig. 5. While we congratulate our BIPM colleagues for making cells on a consistent basis, it is clear that the capability of absolute-frequency measurement referenced to the Cs standard gives us a new approach to characterize reference cells and track their long-term variations.

4 Conclusions

A femtosecond-laser-based optical comb spanning an octave bandwidth is employed conveniently to check systematics and measure absolute frequencies of optical standards. We determine the transition frequency of the length standard at 514.67 nm, i.e. $^{127}I_2$ P(13) 43-0 a_3, to be 71.8 ± 1.5 kHz above the recommended value of $582\,490\,603\,370$ kHz. The measurement was made with the cell cold-finger temperature at $-5\,°C$ (as specified in [1]) and at optical power of 1–4 mW for a pump-beam size of 3 mm in diameter. The average value is obtained over a 15-day measurement period. This result is well within the 1-sigma uncertainty (150 kHz) of the recommended value. The pressure-shift coefficient is found to be $-2.5(0.5)$ kHz/Pa.

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