# Stabilization and Frequency Measurement of the $I_2$-Stabilized Nd : YAG Laser

John L. Hall, Long-Sheng Ma, Matthew Taubman, Bruce Tiemann, Feng-Lei Hong, Olivier Pfister, and Jun Ye

***Abstract*— We report improved stabilization results for and progress toward a more accurate frequency measurement of the 532 nm iodine-stabilized system based on a frequency-doubled Nd : YAG ring laser. We confirm the CCL-adopted frequency well within its stated uncertainty (±40 kHz).**

***Index Terms*— Laser stabilization, modulation transfer, Nd : YAG laser, optical frequency measurement.**

## I. Introduction

THERE are a number of attractive metrological features of the green 532 nm light obtained by frequency-doubling the output of the commercially available stable Nd : YAG ring lasers. These advantages include high visibility, exceptionally low intrinsic frequency and amplitude noise levels, and the flexibility to realize various power levels as needed from submilliwatts to literally tens of watts. Because of the strong absorption and narrow linewidth of the $I_2$ lines at 532 nm, it was realized early on that they form an almost ideal stabilization medium to complement the diode-pumped stable Nd : YAG laser. By now, groups at Stanford University [1] and JILA [2]–[4] have investigated a number of these features, including hyperfine structures, frequency intervals between several absorption lines [5], the absolute optical frequency, as well as some details of building an optical frequency standard based on this system. The 1997 meeting of Consultative Committee for Length (CCL) led to a recommended value for the optical frequency of one particular component, $a_{10}$, of the R(56) 32-0 transition. A number of metrology groups have built stabilized laser systems based on these hyperfine resonances, and recently we have participated [6] in the first international intercomparison with the portable system "Y1" of the National Research Laboratory for Metrology (NRLM).

Manuscript received July 2, 1998. This work was supported in part by the National Institute of Standards and Technology under its program of fundamental research into metrological standards, and by the ONR, the AFOSR, and the National Science Foundation. F.-L. Hong was supported by the NRLM.

J. L. Hall is with the Quantum Physics Division, National Institute of Standards and Technology, Boulder, CO 80309-0440 USA.

L.-S. Ma is with the Quantum Optics Laboratory, East China Normal University, Shanghai, China.

M. Taubman and B. Tiemann are with JILA, University of Colorado, and the National Institute of Standards and Technology, Boulder, CO 80309-0440 USA.

O. Pfister is with the Physics Department, Duke University, Durham, NC 27708 USA.

J. Ye is with the Quantum Optics Group, California Polytechnic University, Pasadena, CA 91125.

Publisher Item Identifier S 0018-9456(99)02879-X.

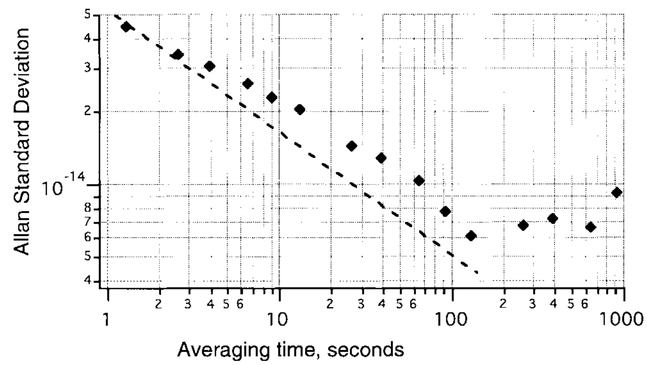



Fig. 1. Frequency stability of frequency-doubled Nd : YAG laser stabilized to iodine transition at 532 nm using modulation-transfer spectroscopy. Calculated short-term performance is approximated at short time, but variations of systematic offsets from the EOM limit performance for times ~30 s and beyond. These contemporary experiments with active control of residual amplitude modulation appear to offer promising advances of long-term stability and reproducibility.

## II. Improved Frequency Stabilization

The large molecular weight of $^{127}I_2$ leads to a low most probable thermal velocity of 140 m/s. The corresponding (fractional) second-order Doppler shift is only $1.1 \times 10^{-13}$, about the same as that of $OsO_4$, the present champion "optical" frequency reference system [7]. Even for the case of ±4.9 K temperature fluctuations, unusual in a modern metrology lab, the corresponding change in the quadratic Doppler shift still is only ±1 Hz in the green. At the iodine operating pressure of 0.5 Pa (−18 °C sidearm temperature), a 1 °C temperature increase gives a frequency decrease of ~225 Hz, a shift rate of −3.2 kHz/Pa. Interestingly, the JILA and NRLM systems have intensity shifts of similar magnitudes, ~2 Hz/$\mu$W, but opposite signs, suggesting that wavefront curvature may play an important role. The use of modulation transfer spectroscopy [8] should effectively suppress any additive offset due to spurious amplitude modulation (AM) of the locking system's phase modulator. Unfortunately, there are in fact some weak "molecular grating reflection" terms that do reflect some of the saturating beam, including its weak AM parts, back toward the detector. Recent improvements in modulator techniques are reducing these problems usefully, which can be judged from the unprecedented low flicker level of our stabilized system, as shown in Fig. 1. This data also shows the nice value $5\times10^{-14}$ stability at 1 s obtained by a first-pass optimization of the system parameters. Apart from some coherent frequency modulations due to a remediable cause (inadequate thermal stability of an unfortunately parallel window on one cell), the Allan deviation averages down tenfold by 100 s to $\sim5\times10^{-15}$, where it intersects the present flicker level of $\pm 2.8$ Hz, about $5 \times 10^{-6}$ of the line's FWHM. We are optimistic that significant further advances are possible, despite the unprecedented low level already demonstrated here. Servo control of the residual amplitude modulation should be an effective tool. In particular, the cavity methods studied previously [9] will be helpful in increasing the signal contrast, even if the remarkable sensitivity [10] of the full noise-immune cavity-enhanced optical heterodyne molecular spectroscopy (NICE-OHMS) approach is not needed.

## III. Frequency Measurement: Principle, Overview, and a Few Details

Our frequency measurement method is based on the frequency interval bisection idea of [11], and creates a stabilized diode laser frequency at 632.109 9 nm. This is the midpoint between the 532 nm radiation of interest and the very effective secondary standard at 778 nm, based on Doppler-free, two-photon absorption in Rb atoms [12]. It is a remarkable accident that the optical frequency of this midpoint is only 660.5 GHz above the customary HeNe-based, Iodine-stabilized reference at 633 nm. This interval is readily spanned with Kurogi's optical frequency comb generator (OFCG) technique [13]. We are now using auxiliary cavity mirrors at both ends of the OFCG. At the input, a power-recycling input mirror [14] serves to vastly reduce the reflection of the cavity's input mirror by forming a transmission resonance at (only) the input optical frequency (from the 633 nm reference system). The output mirror of the OFCG consists of a closely spaced doublet [15] that passes only the desired optical frequency sideband order, the sixty-third in the present case. This comb generator's output beam is heterodyned with a portion of the diode laser's 632 nm output, giving at present some 49 dB SNR in a 100 kHz bandwidth (BW), which approaches sufficiency for direct counting. By contrast, the UV beat is weaker, $\sim$15 dB SNR in the same BW, and requires a tracking oscillator. In fact, both beats are regenerated with this method. The 778 nm beam ($\sim$4 mW) and the 532 nm ($\sim$15 mW) are summed (single-passed) in an angle-matched crystal ($LiIO_3$) while the $\sim$1 mW diode light at 632 nm is doubled in a ring cavity using rubidium di-hydrogen phosphate (RDP) heated to the phase-match temperature near 46 °C. Both UV beams are in the (1–10) nW range so the UV beat is readily obtained with a miniature fast-response photo multiplier tube (PMT). Since the diode laser is narrowed to $<1$ kHz linewidth by fast electronic locking onto a stable reference cavity, the phase-locked tracking oscillators can have a rather leisurely attack rate, $\sim$40 kHz. When the system is running stably, we can confirm that the UV beat is counting reliably by monitoring the limited range of time-domain phase variations of the noisy input wave on an oscilloscope triggered by the regenerated digital wave. When locked, phase variations well below $\pi/5$ rad rms are observed. This comfortable range is confirmed also in the balanced mixer/phase-detector output error signal. Another diagnostic is the $1/\sqrt{\tau}$ averaging of the beat's frequency variation, with a coefficient equal to that of the HeNe/$I_2$ reference laser.

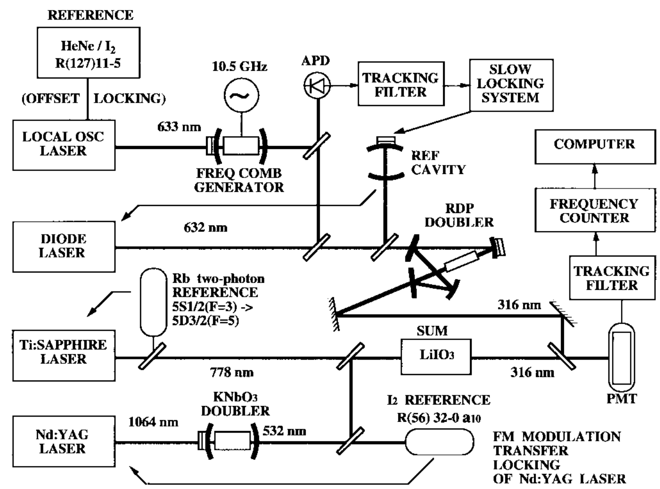



Fig. 2. Block diagram of the frequency measurement system. The comb generator uses an auxiliary mirror on the input for improved coupling efficiency, and on the output to select the single comb line of interest, number +63. The so-formed optical frequency shifter has a "transmission" of about 10% for the +660 GHz output comb line.

## IV. Optoelectronic Configuration

Fig. 2 shows the layout of our system. The Ti : Sapphire laser and the Rb two-photon spectrometer are located on their own (connected) tables, while the JILA pair of Nd:YAG/iodine spectrometers share a table in another part of the lab. The red HeNe reference laser (Winters EO Model 100), the offset laser (1 mW HeNe), and the tunable diode laser are located on a third large table, along with the frequency doubling and summing systems, the comb generator system, and the several heterodyne detectors. Two LambdaMeters are used to control the diode and Ti : Sapphire lasers' wavelengths. Four monomode fibers are used for transporting the beams between tables for the frequency summing or for the LambdaMeter connections.

The operating conditions of the 532 nm spectrometer are presented in [5]. The Rb two-photon spectrometer uses EOM phase modulation to provide a 2.5 rad peak at 100 kHz, and synchronously detecting the fluorescence at this frequency to recover a "discriminator" signal, which is integrated and used to slowly guide the frequency of the prestabilized Ti : Sapphire laser. Basically, a voltage-controlled-oscillator (VCO) receives the Rb error correction and sets the frequency of a large-bandwidth acoustooptic modulator (AOM) to shift the Ti : Sapphire laser slightly relative to its stable reference cavity. In this way the induced frequency noise was below 2 kHz in a 1 s averaging time. For the two-photon spectroscopy, the 8 mW IR input power, (used in a $w_0 = 0.5$ mm beam without a buildup cavity) should generate an ac Stark shift of about $-2$ kHz, according to the data of [12].

Since the 1.3 THz optical offset, measured here as $2 \times 660.5$ GHz, is already about 0.2% of the optical frequency, it is necessary to provide an accurate reference for the RF synthesizer, which drives the comb generator at 10.5 GHz. In this work, a very quiet 10 MHz quartz oscillator was used for that purpose, but its frequency was low by 4.2 parts in $10^9$, as determined by tracking GPS-generated time marks averaged over a few days. Since it was convenient to calculate the output data on the basis of the synthesizer's nominal output frequency, our data appear at frequencies too high by $2 \times 4.2 \times 10^{-9} \times 660.5$ GHz $\cong 5.6$ kHz. This can be seen from our measurement principle, which may be expressed essentially by

$$f(532\text{ nm}) + f(778\text{ nm}) = 2 \times (f(633\text{ nm}) + 660.5\text{ GHz}) + \text{UV beat.} \qquad (1)$$

This leads to

$$f(532\text{ nm}) = 2 \times (f(633\text{ nm}) + 660.5\text{ GHz}) - f(778\text{ nm}) + \text{UV beat.} \qquad (2)$$

So, remembering also the Rb light shift offset, the as-measured/plotted frequencies will need to be corrected by $(-5.6 + 2)$ kHz to take these two offsets into account.

## V. Frequency Measurement Results

All aspects of this measurement have operated independently and well at this time, although the system's continuous running time is not yet ideal. The most critical part of the system seems to be the output filter cavity (finesse $=$ 600) of the comb generator: this resonant output-coupler approach leads to a huge improvement in SNR compared with a simple comb system, but this is true only when its passband is tuned to transmit the sideband of interest. A (small) dither lock to maximize the RF beat power should keep this piezoelectric transducer (PZT) tuning correct for an extended time (hours), whereas mere temperature stabilization seldom provided more than 15 min without compromised signals. A great convenience is the nice operation of an auxiliary servo system that measures the red beat (between the diode and the sixty-third sideband of the HeNe laser) and controls it to be exactly 25 times a crystal reference frequency, 1.0 or 1.1 MHz in different runs. The Allan deviation of this control beat is below $1 \times 10^{-14}$ for 3 s gate times. So for the frequency measurement we only need to measure the UV beat versus time and note the experimental conditions such as synthesizer settings.

The present status of our preliminary measurements is summarized in Fig. 3. The first set of measurements on April 16, 1998, shows a small scatter, near just that expected for the HeNe reference laser performance. The data of April 18, 1998, are seen to be shifted by +15 kHz, which was associated with an integrator offset of 10 mV that was accidentally imposed on the HeNe system during this run. (Perhaps the stability was compromised somewhat as well.) As may be seen from the line in Fig. 3, these data, after this correction, are fully consistent with the earlier run. This shift is just twice that calculated from the measured discriminator slope of 750 Hz/mV, as expected since we double this laser's output frequency. The data of June 8, 15, and 17 are included only to indicate the range of variations possible when the measuring system is tormented: tests include varying the match of the bandpass noise filter with the beat frequency, and changing the gain in the phase-tracking oscillators for the red and UV.

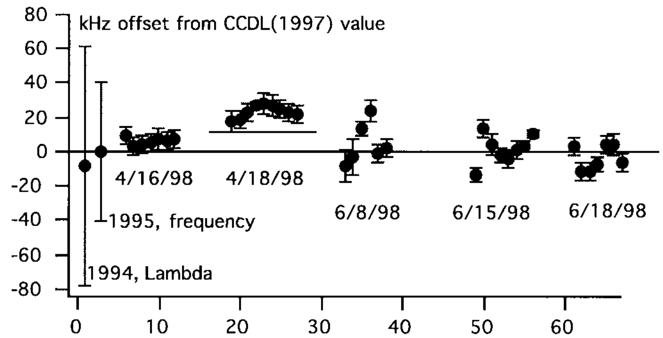



Fig. 3. Summary of preliminary frequency measurements, plotted as frequency offset from the CCL (1997) value, $f(a_{10}) = 563, 260, 223, 471$ kHz. All 1998 data subject to corrections discussed in text, totalling ($-1.6 \pm 10$) kHz. Data of 4/18/98 shifted by 15 kHz associated with zero offset of HeNe reference, as shown. All June data taken as diagnostic measurements, with mismatch of beat and RF filter center frequency, incorrect gain settings of tracking VCO systems, deliberately reduced beat SNR, low beat signal size, etc. The data of this figure makes it sure that the CCDL frequency recommendation is satisfactory, but further systematics tests are underway.

## VI. Conclusions and Accuracy Expectations for the Future

From Fig. 3, it is clear we are not quite ready to announce a new and improved value for the absolute frequency of the Iodine-stabilized Nd : YAG laser. However, equally it is clear that the recommended value of the 1997 CCL is completely sound. As for the future possibilities, the intrinsic uncertainty expected from our measurement system is not more than a few hundred hertz. The published Rb two-photon standard is known to $\pm 2$ kHz, while the physical Rb systems at the Laboratoire Primaire du Temps et des Fréquences (LPTF) are known to about 1 kHz [16]. So our Rb 778 nm standard realization should be confirmed by transporting a system to Paris for heterodyne comparison. The $\pm 12$ kHz nominal uncertainty of the HeNe/$I_2$ system was reduced by the 1997 NORAMET Intercomparison in Querétaro, Mexico, where the JILA laser was found to be at ($-2.3 \pm 1.9$) kHz relative to the "as-maintained" BIPM-4 laser.[1] Unfortunately, use of our laser in the intervening time has led to a lower output power, 45 $\mu$W instead of 70 $\mu$W, and so increased the frequency by ($3.0 \pm 0.5$) kHz. Although our confidence in this frequency is somewhat weakened, for this interim report we adopt ($+1 \pm 5$) kHz as the offset frequency, which appears doubled in (1) and (2), leading to a total green correction of $(-5.6 + 2 + 2 \times 1)$ kHz $= -(1.6 \pm 10)$ kHz.

However, the HeNe reference's uncertainty soon will be reduced to the Rb-system's level by replacing the HeNe with the summed output from the Rb system and a transportable and calibrated methane-stabilized laser to be provided by Prof. M. Gubin of the Lebedev Laboratory: it is another remarkable accident that Rb $+$ $CH_4$ $=$ $I_2$/HeNe, suitably interpreted, to within 50 GHz. Thus, within perhaps one year, the frequency of the exceedingly stable green 532 nm laser light should be known at a level well below 2 kHz.

[1] This value is corrected to the CCL-recommended intracavity power of 10 mW (70 $\mu$W output).

Indeed, in view of the simplicity and extremely attractive performance of the Iodine-stabilized Nd : YAG laser system, at NIST we are seriously considering to building up a full microwave-to-optical synthesis system, using the Hänsch–Meschede–Telle [11] optical frequency-interval-splitting concept ("Divide and Conquer!"). The scheme is made feasible by practical diode lasers, "designer" nonlinear crystals such as periodically poled lithium niobate and CDA, and improvements in Kurogi's optical comb generator method [13] to drastically reduce the required number of optical stages. One attractive scheme focuses around 850 nm and uses $KNbO_3$ for the nonlinear crystals. Another interesting chain converges to 1064 nm, where the comb output could also serve to measure the Nd : FAP laser at 1126 nm, which is used for the trapped-$Hg^+$ optical clock transition.

## ACKNOWLEDGMENT

Helpful discussions with F. Walls, L. Hollberg, and their colleagues at NIST are greatly appreciated by the authors.

## REFERENCES

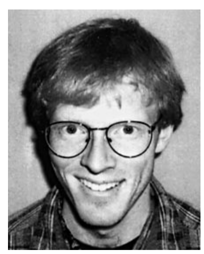
[1] A. Arie, E. K. Gustafson, and R. L. Byer, "Absolute frequency stabilization of diode-pumped Nd : YAG lasers to hyperfine transitions in molecular iodine," *Opt. Lett.*, vol. 17, pp. 1204–1206, Sept. 1992.
[2] M. L. Eickhoff and J. L. Hall, "Optical frequency standard at 532 nm," *IEEE Trans. Instrum. Meas.*, vol. 44, pp. 155–158, Apr. 1995.
[3] P. A. Jungner, S. Swarz, M. L. Eickhoff, J. Ye, J. L. Hall, and S. Waltman, "Absolute frequency of the molecular iodine transition R(56) 32-0 near 532 nm," *IEEE Trans. Instrum. Meas.*, vol. 44, pp. 151–154, Apr. 1995.
[4] P. Jungner, M. L. Eickhoff, S. D. Swarz, J. Ye, and J. L. Hall, "Stability and absolute frequency of molecular iodine transitions near 532 nm," *Proc. SPIE*, vol. 2378, pp. 22–34, 1995.
[5] J. Ye, L. Robertsson, S. Picard, L.-S. Ma, and J. L. Hall, "Absolute frequency atlas of molecular $I_2$ lines at 532 nm," this issue, pp. 544–549.
[6] F.-L. Hong, J. Ishikawa, J. Yoda, J. Ye, L.-S. Ma, and J. L. Hall, "Frequency comparison of $^{127}I_2$-stabilized Nd:YAG lasers," this issue, pp. 532–536.
[7] A. Clairon, O. Acef, C. Chardonnet, and C. J. Bordé, "State of the art for high accuracy frequency standards in the 28 Thz range using saturated absorption resonances of $OsO_4$ and $CO_2$," in *Frequency Standards and Metrology*, A. deMarci, Ed. Berlin, Germany: Springer-Verlag, 1989, pp. 212–221.
[8] J. H. Shirley, "Modulation transfer processes in optical heterodyne saturation spectroscopy," *Opt. Lett.*, vol. 7, pp. 537–539, 1982.
[9] L.-S. Ma and J. L. Hall, "Optical heterodyne spectroscopy enhanced by an external cavity: Toward improved working standards," *IEEE J. Quantum Electron.*, vol. QE-26, pp. 2006–2012, 1990.
[10] J. Ye, L.-S. Ma, and J. L. Hall, "Ultrasensitive detections in atomic and molecular physics: Demonstration in molecular overtone spectroscopy," *J. Opt. Soc. Amer.*, vol. B15, pp. 6–15, 1998.
[11] H. R. Telle, D. Meschede, and T. W. Hsnsch, "Realization of a new concept for visible frequency division: Phase-locking of harmonic and sum frequencies," *Opt. Lett.*, vol. 10, pp. 532–534, 1990.
[12] D. Touahri, O. Acef, A. Clairon, J.-J. Zondy, R. Felder, L. Hilico, B. de Beauvoir, F. Biraben, and F. Nez, "Frequency measurement of $5S(F=3)-5D(F=5)$ two-photon transition in Rb," *Opt. Commun.*, vol. 133, pp. 471–478, 1997, and references therein.
[13] M. Kourogi, K. Nakagawa, and M. Ohtsu, "Wide-span optical frequency comb generator," *IEEE J. Quantum Electron.*, vol. 29, pp. 2693–2701, 1993.
[14] M. Kourogi, T. Enami, and M. Ohtsu, "A coupled-cavity monolithic optical frequency comb generator," *IEEE Photon. Technol. Lett.* vol. 8, pp. 1698–1700, 1996
[15] A. S. Bell, G. M. Mcfarlane, E. Riis, and A. I. Ferguson, "Efficient optical frequency-comb generator," *Opt. Lett.*, vol. 12, pp. 1435–1437, 1995.
[16] J. Ye, L. S. Ma, T. Day, and J. L. Hall, "Highly selective terahertz optical frequency comb generator," *Opt. Lett.*, vol. 22, p. 301, 1997.
[17] O. Acef, Laboratoire Primaire du Temps et des Fréquences, private communication, Paris, France, July 1998.

**John L. Hall**, for a photograph and biography, see this issue, p. 536.

**Long-Sheng Ma**, for a photograph and biography, see this issue, p. 536.

**Matthew Taubman** was born on August 19, 1966, in Young, NSW, Australia. He received the B.Sc. Hon. degree (with the University Medal in physics) in 1989 and the Ph.D. degree in physics for research in experimental quantum optics in 1995, both from the Australian National University, Canberra.

From October 1995 to April 1997, he was a Postdoctoral Fellow, Virgo Gravitational Wave Interferometer Group, Orsay, France, working on power stabilization of the laser system to be used in the Virgo Interferometer. Since that time, he has worked with Dr. John Hall at the University of Colorado, Boulder, as a Postdoctoral Fellow, where his research includes laser stabilization, frequency standards, frequency locking of laser/cavity and laser/molecular systems, and the application and refinement of control systems for precision measurement.



**Bruce Tiemann** was born in Schenectady, NY, in 1965. He received the B.S. degree in chemistry from the California Institute of Technology, Pasadena, in 1987. He is presently in the graduate program at the University of Colorado, Boulder.

He has held a staff position at the California Institute of Technology, working on organic molecules for nonlinear optics with Seth Marder of the Beckman Institute.

**Feng-Lei Hong**, for a photograph and biography, see this issue, p. 535.

**Olivier Pfister** was born on August 12, 1964, in Nice, France. He received the B.S. degree in 1987 from the University of Nice and the M.S. and Ph.D. degrees in 1989 and 1993, respectively, from the University of Paris-Nord, Paris, France.

He was a Postdoctoral Researcher at JILA, the University of Colorado, and the National Institute of Standards and Technology, Boulder, CO, from 1994 to 1997. He is currently a Research Associate in the Quantum Electronics Group, Department of Physics, Duke University, Durham, NC. His research interests include quantum-optics with multiphoton lasers and oscillators, ultrasensitive interferometry, nonlinear optics for frequency standards, ultrastable lasers, and ultrahigh-resolution molecular spectroscopy.

Dr. Pfister is a member of the American Physical Society and of the Optical Society of America.

**Jun Ye**, for a photograph and biography, see this issue, p. 536.