Direct frequency measurements of transitions at 520 THz (576 nm) in iodine and 260 THz (1.15 μm) in neon

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Received November 2, 1982

The *o* hyperfine component of the $^{127}I_2$ 17-1 $P(62)$ transition at 520 THz (576 nm) in iodine was measured with respect to the CH_4-stabilized 88-THz He–Ne laser. A 26-THz CO_2 laser, a color-center laser at 130 THz, and a He–Ne laser at 260 THz were used as transfer oscillators. The measured I_2 frequency was 520 206 808.547 MHz with a total fractional uncertainty of 1.6×10^{-10}. The 1.15-μm ^{20}Ne Lamb-dip-stabilized laser frequency was 260 103 249.26 MHz with a total fractional uncertainty of 3.1×10^{-10}.

There is at present considerable interest in redefining the meter in terms of the second. The Comité Consultatif pour la Définition du Mètre has proposed: "The meter is the length of the path travelled by light in vacuum during a time interval of 1/299 792 458 of a second."[1] With this definition, the meter could be realized from the wavelength of *any* laser that is stabilized to a narrow atomic or molecular absorption for which the frequency is known. The wavelength λ would be determined from the relation $\lambda = c/\nu$, where c is the fixed value of the speed of light and ν is the measured frequency of the laser. In practice, to realize this definition it will be necessary to measure the absolute frequency of suitable spectral lines in the visible spectral region. Before the frequency measurements reported here were made, accuracies better than that of the present Kr length standard (4 parts in 10^9) have been limited to various infrared lasers, such as the CH_4-stabilized He–Ne laser[2–5] and the saturated-absorption-stabilized CO_2 laser.[3] In this Letter, we report the cw synthesis and measurement of the frequency of the *o* hyperfine component of the visible $^{127}I_2$ 17-1 $P(62)$ transition at 520 THz (576 nm) to 1.6 parts in 10^{10} and of the Lamb-dip-stabilized ^{20}Ne laser at 260 THz (1.15 μm) to an accuracy of 3.1 parts in 10^{10}. This accuracy represents an improvement by nearly 3 orders of magnitude over previous measurements.[6] (The accompanying Letter reports the absolute frequency measurement of the I_2-stabilized 633-nm He–Ne laser with comparable accuracy.[7]) The frequencies were measured by comparing them with the known frequency of the 11.515-μm $^{13}C^{16}O_2$ laser (referenced to the CH_4-stabilized He–Ne laser) using sixth-order mixing in a point-contact diode and with subsequent use of a 2.3-μm color-center laser and a 1.15-μm He–Ne laser as frequency-transfer oscillators.

The laser-frequency synthesis chain is shown in Fig. 1. In this chain, the sum of three harmonics of one CO_2 laser plus two harmonics of another are heterodyned with the 130-THz color-center laser radiation on a W–Ni point-contact metal–insulator–metal (MIM) diode. Because the MIM diode becomes less efficient with increasing frequency, it has not been possible to date to generate more than five harmonics of CO_2 laser radiation in the diode. Therefore transfer oscillators at 2.3 and 1.15 μm in conjunction with nonlinear frequency-doubling crystals are used to extend the measurement to the visible. Part of the color-center radiation is thus frequency doubled and heterodyned with the 260-THz (1.15-μm) He–Ne laser on a Ge photo-

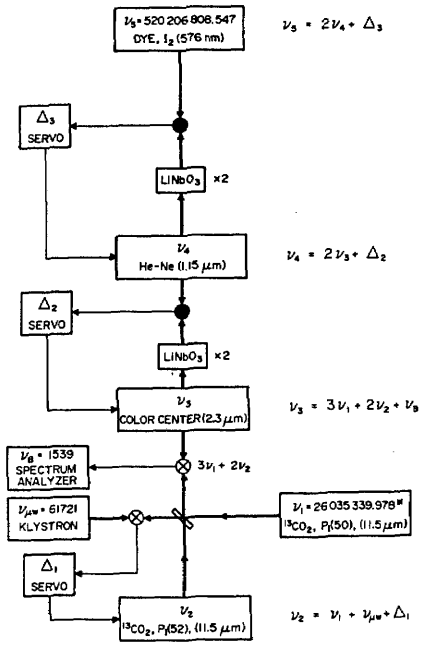

Fig. 1. At the top of the chain, a cw dye laser is locked to an I_2 transition. A 1.15-μm He–Ne laser is locked to the dye laser, and a 2.3-μm color-center laser is locked to the He–Ne laser. The beat between the CO_2 lasers and the color-center laser is measured.

0146-9592/83/030133-03$1.00/0

diode. The 260-THz He–Ne laser radiation is then frequency doubled and heterodyned with the output of a 520-THz (576-nm) cw laser frequency locked to an I_2 line observed in saturated fluorescence.

The CO_2 laser radiation was generated by two $^{13}C^{16}O_2$ lasers operating on the adjacent lines $P_I(50)$ and $P_I(52)$. These CO_2 lines were selected because twice the frequency of one CO_2 laser plus three times the frequency of the other nearly equaled the necessary color-center laser frequency. The $P_I(50)$ laser was offset locked to a second $P_I(50)$ laser, which was stabilized by using the saturated fluorescence signal at 4.3 μm from an external $^{13}C^{16}O_2$ absorption cell filled to 5.3 Pa. The $P_I(52)$ laser was phase locked to the $P_I(50)$ laser by a stabilized 62-GHz klystron. The two CO_2 laser beams were apertured for individual power control and combined collinearly with a beam splitter.

The combined CO_2 radiation (approximately 200 mW) was focused at 45° with respect to the whisker onto a W–Ni point-contact MIM diode. Coincident upon the diode at −45° was 10 mW of 2.3-μm radiation from the color-center laser. The beat generated on the MIM diode (ν_B in Fig. 1) was displayed on an rf spectrum analyzer along with frequency markers from a counted rf oscillator. The signal was averaged for 30 sec on a signal averager and plotted for later interpolation between the two known frequency markers. The averaged rf beat was typically 50 kHz wide (FWHM).

The 2.3-μm cw color-center ring laser was used as a frequency-transfer oscillator for connecting the CO_2 lasers to the 1.15-μm He–Ne laser. The color-center laser used $(F_2^+)_A$ centers in Li-doped KCl and provided broadly tunable laser output from 2.0 to 2.5 μm.[8] When pumped with 4 W of power from a cw 1.3-μm Nd:YAG laser, the color-center laser produced 120 mW of single-mode output power at 2.3 μm. The laser was operated in a ring configuration[9] to ensure efficient, single-mode operation and was actively stabilized to a stable optical resonator with a resulting linewidth of approximately 10 kHz. The 2.3-μm radiation from the color-center laser was focused into a temperature-tuned, 90° phase-matched $LiNbO_3$ frequency-doubling crystal (T = 530°C; dimensions 4 mm × 4 mm × 50 mm) (Ref. 10) and generated approximately 10 μW of cw 1.15-μm radiation. This second-harmonic radiation was combined on a beam splitter with 10 mW of radiation from a 1.15-μm He–Ne laser, and both beams were then focused onto a Ge photodiode. The subsequent rf beat, Δ_2, was displayed on an rf spectrum analyzer in a manner similar to the CO_2 laser color-center laser beat and was also used to frequency lock the color-center laser to the 1.15-μm laser.

The He–Ne 260-THz laser employed an 8-m discharge tube operating at 800-Pa total pressure. This length of laser ensured sufficient power for mixing and frequency doubling. A resonant reflector consisting of a 99% reflective end mirror and a thin lossy metallic film on a piezoelectric transducer (PZT) positioned 9 cm from the end mirror provided tuning and mode selection. A fast PZT on the output coupler was used for servo frequency control. Approximately 90 mW of the output power was focused into a temperature-tuned (T = 185°C) (Ref. 10) 25-mm-long $LiNbO_3$ frequency-doubling crystal to generate 50 μW of 520-THz radiation. This visible second-harmonic radiation was collinearly combined with a portion of an I_2-stabilized cw dye-laser output and focused onto a photodiode to produce an rf beat, Δ_3. This signal was amplified and sent through a discriminator to provide an error voltage for frequency locking the He–Ne laser to the I_2 by means of the dye laser.

At the top of the chain, a 520-THz cw dye laser was servo locked to the hyperfine o component of the $^{127}I_2$ 17-1 $P(62)$ transition. Locking was accomplished by a first-derivative scheme, with the laser modulated at 100 kHz to a depth of 0.5 MHz. The I_2 transition had a linewidth (FWHM) of 1 MHz for a pressure of 4.5 Pa. The o component was positioned closest to the center of the doubled 1.15-μm laser line, and since it was well separated from the other hyperfine components, it had a relatively undistorted line shape. Corrections for systematic offsets of the lock point from true line center are discussed below.

In a separate experiment, the frequency of the Lamb-dip-stabilized ^{20}Ne laser was subsequently measured relative to the I_2 o component. The 8-m He–Ne laser was offset locked 60 MHz from the ^{20}Ne laser, and from the resulting beat between the second harmonic of the He–Ne laser and the I_2-stabilized dye laser it was possible to determine the ^{20}Ne laser frequency.

The results of the frequency measurements using ν_{CH_4} = 88 376 181.609 ± 0.009 MHz are that

$$\nu_{I_2} = 520\,206\,808.547 \pm 0.081 \text{ MHz}$$

and

$$\nu_{Ne} = 260\,103\,249.26 \pm 0.081 \text{ MHz}.$$

These frequencies, which are in agreement with previous less-accurate measurements,[6] represent line-center-value uncertainties (1-σ estimates) of 1.6 parts in 10^{10} for the visibl3 frequency and 3.1 parts in 10^{10} for the ^{20}Ne frequency. Table 1 shows the error budget for the visible measurement. The largest error arises from the uncertainty in the frequency of the CO_2 reference laser at the bottom of the chain. The $P_I(50)$ laser used

Table 1. Error Budget for the Visible-Frequency Measurement

Component	Error (kHz)
CO_2 laser uncertainty (3.4 kHz times effectively 20 harmonics)[a]	68
Electronic resettability of entire I_2 servo system	40
Uncertainty in first-derivative offsets and pressure shifts	15
Statistical fluctuation in data	14
Total uncertainty (summed quadratically)	81

[a] This 3.4 kHz is due to an estimated 1×10^{-10} uncertainty in the CH_4-stabilized He–Ne frequency added in quadrature to the measurement uncertainties in the determination of the $P_I(50)$ frequency.

in this experiment was compared against a CH_4-stabilized He–Ne laser operating at 3.39 μm by use of a point-contact MIM diode and various CO_2 laser lines. The results of this measurement will be reported separately. We note here only that the $P_I(50)$ laser frequency was resettable to within 5 parts in 10^{11}, and since the CH_4-stabilized He–Ne laser frequency is resettable to 3 parts in 10^{11} and is known absolutely to 1 part in 10^{10} (this results from averaging the four most recent international measurements of the CH_4-stabilized He–Ne laser),[2–5] the frequency of the $P_I(50)$ laser was determined to nearly this accuracy. The CO_2 uncertainty (3.4 kHz) was multiplied by 20 in the process of synthesizing the 520-THz frequency.

At the top of the chain, the reproducibility of the I_2 lock point was affected by several parameters. First, we observed a possible 40-kHz shift in the lock point over an eight-week period caused by a misadjustment in the servo electronics. This offset could certainly be reduced or eliminated in a subsequent remeasurement; however, the reduction in total uncertainty would be insignificant because of the comparatively large CO_2 uncertainty. Hence a remeasurement was not undertaken. An offset inherent with first-derivative locking schemes was observed, caused by the servo system's locking to zero voltage instead of locking to the crossing of the hyperfine signal with the Doppler background. The offset correction was measurable in terms of percentage of line height and line width, and a correction was made in the frequency reported above. The modulation width of the dye laser was adjusted to minimize modulation offsets. Reductions below 0.5-MHz modulation width only reduced the signal-to-noise ratio, with an insignificant (less than 5-kHz) shift of the lock point. The temperature of the I_2 was systematically varied over 27°C to determine pressure shifts. Although there are no published data on the I_2 line used in this measurement, the shifts observed were approximately the same as those observed at 633 (Ref. 11) and 612 nm (Ref. 12) on $^{127}I_2$ and $^{129}I_2$, respectively, namely, −7.5 kHz/Pa. Hence the value reported above includes the +31-kHz extrapolation to zero pressure. The corrections for pressure shift and lock-point offset had an uncertainty of 15 kHz.

The uncertainties caused by laser drift during measurements and by interpolation errors appeared as scatter in the data and amounted to 14 kHz. Since all the uncertainties listed in Table 1 are uncorrelated, they were added quadratically to yield an 81-kHz uncertainty, or 1.6 parts in 10^{10}.

The Lamb-dip-stabilized ^{20}Ne laser has been described previously.[6] When measured against the *o* component, it was found to have a 1-σ scatter of 71 kHz. The total uncertainty of the measurement is thus the quadrature sum of 71 and 40 kHz (uncertainty of the I_2 frequence at 1.15 μm) to yield 81 kHz. Several times during the measurements, the laser was grossly misaligned and realigned to ensure that there were no offsets from spatial asymmetry of the mode. No shifts were detected. The laser was dithered at 4 kHz with a 10-MHz peak-to-peak amplitude.

In conclusion, we have measured the absolute frequency of a visible 520-THz (576-nm) I_2 transition to an accuracy of 1.6 parts in 10^{10} and of the 260-THz (1.15-μm) ^{20}Ne Lamb-dip-stabilized laser to 3.1 parts in 10^{10}. This represents an improvement by almost 3 orders of magnitude in the accuracy of frequency measurements in the visible. The accuracy reported approaches the current accuracy with which the frequency of the CH_4-stabilized He–Ne laser is known. Significant improvement in these accuracies will require a remeasurement of methane, narrower spectral features in the visible, and probably a more-elaborate synthesis scheme, such as a phase-locked laser-frequency chain from a visible transition to the atomic-clock time standard, or realization of new schemes, such as the proposed frequency divider.[13]

It is a pleasure to thank J. C. Bergquist and J. L. Hall for many useful discussions about laser stabilization. We are also indebted to the NASA Upper Atmosphere Research Office for partial support in developing some of the lasers used in this experiment.

References

1. Report: Comité Consultatif pour la Définition du Mètre, 7th Session (Bureau International des Poids et Mesures, Sèvres, France, 1982).
2. D. J. Knight, G. J. Edwards, P. R. Pearce, and N. R. Cross, IEEE Trans. Instrum. Meas. **IM-29,** 257 (1980).
3. A. Clairon, B. Dahmani, and J. Rutman, IEEE Trans. Instrum. Meas. **IM-29,** 268 (1980).
4. Bulletin G-16 ISSN No. 0135-2415 Gosstandard (Commission of United Time Service, Moscow, USSR).
5. V. P. Chebotayev, J. Phys. (Paris) **42,** C8-505 (1981).
6. K. M. Baird, K. M. Evenson, G. R. Hanes, D. A. Jennings, and F. R. Petersen, Opt. Lett. **4,** 263 (1979).
7. D. A. Jennings, C. R. Pollock, F. R. Petersen, R. E. Drullinger, K. M. Evenson, J. S. Wells, J. L. Hall, and H. P. Layer, Opt. Lett. **8,** 136 (1983).
8. I. Schneider and C. L. Marquardt, Opt. Lett. **5,** 214 (1980).
9. C. R. Pollock and D. A. Jennings, Appl. Phys. B **28,** 308 (1982).
10. Both temperature-tuned crystals were surrounded by heat pipes (530°C required potassium; 185°C required a fluorocarbon). The temperatures were maintained to within ±0.2°C.
11. J. M. Chartier, J. Helmcke, and A. Wallard, IEEE Trans. Instrum. Meas. **IM-25,** 450 (1976).
12. P. Cerez, A. Brillet, C. N. Man-Pichot, and R. Felder, IEEE Trans. Instrum. Meas. **IM-29,** 352 (1980).
13. D. J. Wineland, J. Appl. Phys. **50,** 2528 (1979).