

The Development of Direct Optical Frequency Measurement and the Speed of Light*

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Since the advent of the laser and saturated absorption locking of the laser, the extension of radio frequency technology into the optical region has produced exciting new results, such as a 10 000-fold increase in the resolution of spectroscopy in this region and a new value of the speed of light, 100 times more accurate than the previously accepted value. This paper will describe the techniques and results achieved.

INTRODUCTION

The advent of coherent stable oscillators (lasers) in the far infrared to optical frequency region has produced many new exciting results. Some of these have resulted from the application of radio techniques (including direct frequency measurements) to this region of the spectrum. In fact, the direct measurement of both the frequency⁽¹⁾ and wavelength⁽²⁾ of the methane stabilized He-Ne laser at 3.39 μm has yielded a value⁽³⁾ of the speed of light which is 100 times more accurate than the previously accepted value. This significant increase in accuracy was made possible by the extension of frequency measurements to the region of the electromagnetic spectrum where wavelength measurements can be made with high accuracy. This new value for the speed of light, which includes an average of wavelength measurements by several laboratories, has recently been modified⁽⁴⁾ by averaging several other wavelength values to obtain the value: 299 792 458.0 m/s $\pm 4 \times 10^{-9}$. It is the purpose

of this paper to describe these direct frequency measurement techniques and their application in the speed of light measurement.

ELECTROMAGNETIC SPECTRUM

The spectral character of an electromagnetic wave is described by either its vacuum wavelength or its frequency (and, of course, the speed of light is the product of the two). Before the advent of lasers, infrared and visible spectra were measured by wavelength techniques. However, lasers have fractional linewidths approaching those of radio and microwave oscillators and have many orders of magnitude more spectral radiance than incoherent sources. Thus, direct frequency measuring techniques as well as wavelength techniques can be used, and the frequency techniques provide much more resolution and accuracy in measuring the spectral characteristics of the radiation. Frequency measuring techniques are limited only by the accuracy of the fundamental standard and the stabilities of the sources available and, as a result, are better by several orders of magnitude. Hence, lasers which provide intense coherent sources of electromagnetic radiation extending from the mi-

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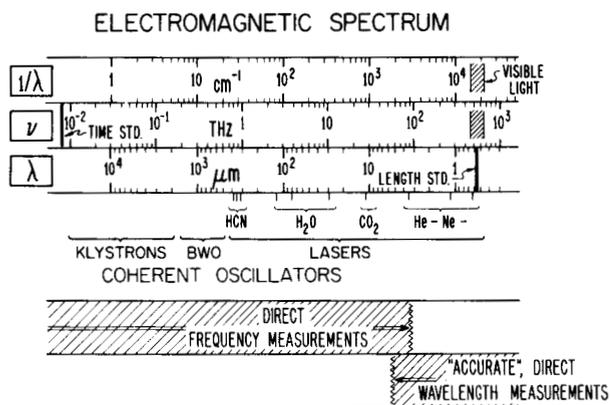


Figure 1. The electromagnetic spectrum showing oscillators, frequency and wavelength standards, and measurement regions. The extension of frequency measurements into the infrared has produced an overlap of the accurate ($< 4 \times 10^{-9}$) wavelength measurement region for the first time. The frequency standard is the zero field hyperfine structure separation in the ground state of ^{133}Cs , which is defined to be 9 192 631 770 Hz. The wavelength standard is the transition between the $2p_{10}$ and $5d_5$ levels of ^{86}Kr with the vacuum wavelength of this radiation defined to be 1/1 650 763.73 m.

crowave through the visible to the ultraviolet portion of the electromagnetic spectrum (see Figure 1) can be considered as either wavelength or frequency sources.

In spite of the fact that lasers provide coherent frequency sources in the infrared and visible, frequencies could not immediately be measured because no device capable of generating frequencies of a few terahertz from harmonics of cw sources was known to exist. Secondly, the laser frequency was not very stable; that is, in the case of the gas laser, although its short-term linewidth was a few hundred hertz, over a long period its frequency could vary within the Doppler and pressure broadened gain curve of the laser. This gain curve might vary from less than a hundred to several thousand megahertz depending on the laser. Therefore, even though the instantaneous frequency of the laser could be measured, there was no reference point other than the broad gain curve whose center could not be located very precisely. Happily, solutions to both of these problems were found: for the first, the extension of frequency measurements into the infrared by means of the metal-on-metal point contact diode,⁽⁵⁾ and for the second, the use of saturated absorption⁽⁶⁾ and optical heterodyne techniques.⁽⁷⁾

The extension of absolute frequency measurements, linking the cesium standards (accurate⁽⁸⁾ to about 2×10^{-13}) to these lasers, provides accuracy as well as resolution to the absolute frequencies involved. At the same time, we use the wavelength aspect of the radiation, for example, in precision long-path interferometry.⁽⁹⁾ Indeed, the increasing resolution

which these new spectroscopic techniques provide may very well usher in an era of precision and accuracy in frequency and length measurements undreamed of a few years ago.

Recent advances in stabilization with saturated absorption techniques have produced lasers with 1 s fractional frequency instabilities⁽⁶⁾ as small as 5×10^{-13} . Although the fractional uncertainty in frequency reproducibility is somewhat larger, stabilized lasers are already excellent secondary frequency standards. With additional development in laser stabilization and infrared frequency synthesis, some of these devices can be considered as contenders for the primary frequency standard role; the $3.39 \mu\text{m}$ He-Ne laser, for example, currently has a fractional frequency uncertainty in reproducibility⁽⁶⁾ of less than one part in 10^{11} , and the 6330 \AA He-Ne laser⁽¹⁰⁾ is reproducible to better than 1×10^{-10} . The duality implies that the radiation must have a similar wavelength characteristic; i.e., $\Delta\lambda/\lambda \approx 10^{-11}$, which is more than 100 times better than the current length standard.⁽⁷⁾ Hence, the stabilized laser must be considered to have tremendous potential in wavelength as well as frequency standards applications and perhaps in both.⁽¹¹⁾

FREQUENCY MEASUREMENTS

Direct frequency measurements require counting the number of cycles of electromagnetic radiation in a given period. Modern electronic circuitry permits direct cycle counting up to about 500 MHz with the timing generally provided by a quartz crystal oscillator, or for the highest accuracy, directly by a cesium clock. A measurement of a higher frequency requires a heterodyne technique in which two oscillators (one whose frequency is known) are mixed together so that a countable frequency difference is generated. The known frequency may be synthesized as an exact harmonic of a directly countable lower frequency by irradiating an appropriate nonlinear device. To synthesize microwave frequencies, oscillators of about a 100 MHz irradiating a silicon diode will generate useful harmonics as high as 40 GHz. The desired harmonic is generally mixed with the unknown frequency in the same diode so that the heterodyne beat note of a few megahertz is finally generated and directly counted.

The above process may be illustrated in the following equation:

$$\nu_B = \nu_x - (l\nu_a) \quad (1)$$

where

$$\begin{aligned} \nu_x &= \text{unknown frequency} \\ l &= \text{harmonic number} \end{aligned}$$

ν_a = reference frequency

ν_B = countable beat note frequency

The harmonic number l is generally determined by making an approximate frequency determination from a wavelength measurement. At frequencies from about 40 to 1000 GHz (1 THz), the silicon diode is still operable; however, several steps are usually required in a frequency measurement chain extending from some countable reference frequency. For this purpose, klystrons between 60 and 80 GHz are often phase-locked to a lower frequency klystron and these are used to cover the range between 200 and 1000 GHz with less than 15 harmonics in each step. The harmonic number is kept below 15 because at frequencies above a few hundred gigahertz the overall harmonic generation-mixing process is less efficient, and consequently one must use a lower harmonic number.

HARMONIC MIXER DIODES

Another type of harmonic-generator mixer, the point contact Josephson junction, is a much more efficient diode in the generation of high harmonics than is the silicon diode. Its use has been extended to 3.8 THz^(12a) by the direct generation of the 401st harmonic from an X-band source. Although this frequency at present is not much higher than that for silicon diode operation, work is continuing on the Josephson junction to extend its high frequency limit.

A third type of harmonic-generator mixer, the meter-metal diode, in either a point contact⁽⁵⁾ or a thin film configuration,^(12b) is the only device which has been used at frequencies above 3.8 THz. Of all the diodes, the metal-metal point contact diode has been the most useful at laser frequencies, and has thus far⁽¹³⁾ operated to 88 THz (3.39 μm). High frequency applications of this diode were first demonstrated in Javan's laboratory⁽⁵⁾ by generating and mixing the third harmonic of a pulsed water vapor laser with a CO₂ laser. Specifically, the tungsten-nickel combination has been more useful than many others which were tested. At these laser frequencies, harmonic generation, sideband generation, and mixing occur in the same diode; that is, the harmonics or sidebands are not actually propagated from diode to diode, but a single diode is irradiated with all of the necessary frequencies. These diodes use a tungsten catwhisker electrochemically pointed to a few hundred angstroms radius at the tip which makes contact with a polished nickel surface. The catwhisker acts as a "long wire antenna"⁽¹⁴⁾ and possesses lobes similar to antennae used at rf frequencies. Laser radiation directed in along a dominant lobe is therefore propagated by the catwhisker to the tungsten-nickel junction.

Some characteristics of this type of diode have been measured⁽¹⁵⁾; and, so far, attempts at extending its range to the visible have shown that it mixes at frequencies as high as 583 THz (5145 Å). However, harmonic generation beyond 88 THz has not been demonstrated. Although the exact physical mechanism by which the harmonic generation mixing occurs is not yet understood, there are at least three different possible explanations for the phenomena. One is a tunneling process through an oxide layer,⁽¹⁶⁾ another is a field emission process,⁽¹⁷⁾ and a third is a quantum mechanical scattering model.⁽¹⁸⁾

As an example of a frequency comparison, one of the steps in measuring the frequency of a CO₂ laser is to compare the frequency of its 9.3 μm R(10) line with the third harmonic of a water vapor laser.

In this case,

$$\nu_{\text{beat}} = \nu_{\text{CO}_2, R(10)} - (3 \nu_{\text{H}_2\text{O}} - \nu_{\text{kly}}) \quad (2)$$

where

$$\begin{aligned} \nu_{\text{CO}_2, R(10)} &= \text{unknown frequency} \\ &(32.134,267 \text{ THz}) \end{aligned} \quad (3)$$

$$\nu_{\text{H}_2\text{O}} = 10.718068 \text{ THz} \quad (4)$$

$$\nu_{\text{kly}} = 0.019958 \text{ THz} \quad (5)$$

$$\nu_{\text{beat}} = 0.000020 \text{ THz} \quad (6)$$

In this heterodyning, the radiation from all three sources simultaneously irradiates the diode and the resulting 20 MHz beat note is amplified and measured in a counter or spectrum analyzer.

The measurement of a beat between two laser plus other oscillators yields the frequency of one laser with respect to another. Due to the inefficiency of the diode at mixing orders higher than about the 15th, a whole chain of lasers and diodes must be used. The speed of light measurement and the chain used will now be described.

THE SPEED OF LIGHT

The speed of light, c , is possibly the most important of all the fundamental constants.⁽¹⁹⁾ It enters into the conversion between electrostatic and electromagnetic units, it relates the mass of a particle to its energy in the well known equation $E = mc^2$, and it is used as well in many relationships connecting other physical constants. In ranging measurements, very accurately measured transit times for electromagnetic waves are converted to distance by multiply-

ing by the speed of light. Examples are geophysical distance measurements such as microwave planetary radar and laser lunar ranging.⁽²⁰⁾ Recent experiments have set very restrictive limits on any possible speed dependence on direction⁽²¹⁾ and frequency.⁽²²⁾

Because of its importance, more time and effort have been devoted to the measurement of c than any other fundamental constant. In spite of this fact, a highly accurate result has remained elusive even in recent times—mainly because of ever present systematic errors which have been difficult to properly evaluate. Indeed, the probable value of c has changed, and the suggestion has been made that the real value may be changing linearly or even periodically with time.⁽²³⁾ However, in view of recent electro-optical measurements, microwave interferometer measurements, spectroscopic measurements, and the laser measurement to be described here, it appears that the probable value of c is converging to a constant within the stated errors and that the changing nature of c in the past was related to unaccounted inaccuracy in previous experiments.

Measurements of c in which both the wavelength and frequency of an oscillator were measured played a significant role in proving that radio and light radiation were electromagnetic in nature. This method was used by Froome⁽²⁴⁾ to obtain the accepted value of c in use since 1958. His method used a moving reflector microwave interferometer operating at 72 GHz which was used to measure the microwave wavelength in terms of the length standard. The vacuum wavelength thus obtained when corrected for diffraction and other systematic effects was multiplied by the measured frequency to give a value for the speed of light. The frequency could be related to the primary standard with good accuracy, but the major estimated experimental error was connected with uncertainties in the wavelength measurement. At these long wavelengths the diffraction problem forced the use of a long air path which resulted in uncertainties in the measurement of the path length and in the index of refraction.

Speed of light determinations from wavelength and frequency measurements have traditionally suffered from the problems illustrated by Froome's experiment. To measure the frequency, it is best to do the experiment at a frequency not too far removed from the primary Cs frequency standard, where extremely stable oscillators can be made and where frequencies are easily measurable. However, to measure the wavelength, it is best to do the experiment close to the visible ⁸⁶Kr wavelength standard, where wavelengths can be more easily compared and where diffraction problems are not as severe. The extension of frequency measurements into the infrared portion

of the electromagnetic spectrum has in a sense solved this dilemma, and has been responsible⁽²⁾ for the 100-fold increase in the accuracy in the value of c . The frequency of the methane stabilized helium-neon laser is over 1000 times higher than that of the oscillator used in Froome's measurement of c . Direct frequency measurements were recently extended to this frequency⁽¹³⁾ and subsequently refined⁽¹⁾ to the present accuracy of 6 parts in 10^{10} . The wavelength of this stabilized laser has been compared^(2,25-27) with the krypton-86 length standard to the limit of the usefulness of the length standard (approximately 4 parts in 10^9).⁽⁴⁾

Methane Frequency Measurement

To measure the frequency of methane, a chain of oscillators extending from the cesium frequency standard to the methane stabilized helium-neon laser was used. The chain is shown in Figure 2. The three saturated-absorption-stabilized lasers are shown in the top of the right-hand section (the methane stabilized laser is in bold outline in the upper right), the transfer chain oscillators are in the center column, and the cesium frequency standard is in bold outline

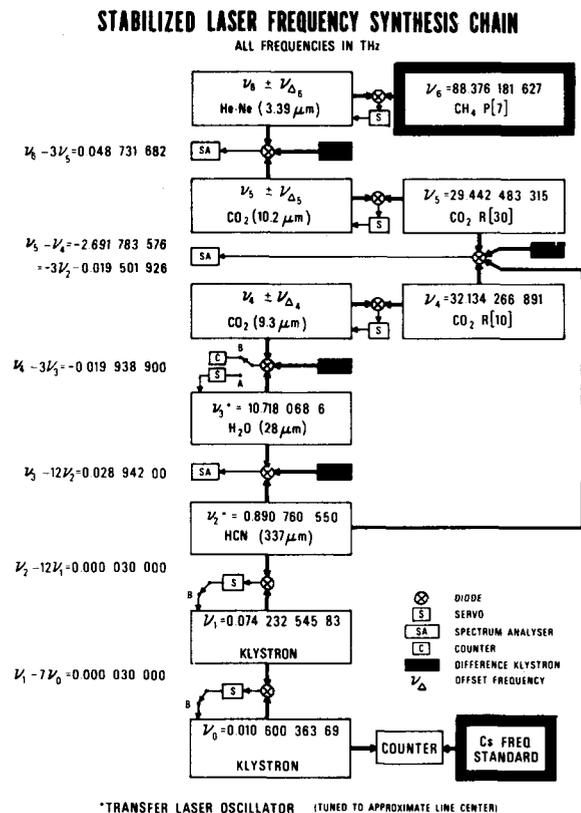


Figure 2. Stabilized laser frequency synthesis chain. All frequencies are given in THz; those marked with an asterisk were measured with a transfer laser oscillator tuned to approximate line center.

in the lower right-hand corner. The chain can be called the cesium to methane frequency synthesis chain. The exact details of the experiment have already been described⁽¹⁾ and will be omitted here.

The methane-stabilized He-Ne laser used in these experiments is quite similar in size and construction to the device described by Barger and Hall.⁽²⁾

The frequencies measured in this chain are

Molecule	Line	λ	Frequency
¹² C ¹⁶ O ₂	R(30)	10.18 μ m	29.442 315 (25) Thz
¹² C ¹⁶ O ₂	R(10)	R99.33	32.134 266 891 (24)
¹² CH ₄	P(7)	3.39	88.376 181 627 (50)

The numbers in parentheses are 1-standard-deviation-type errors indicating uncertainties in the last two digits.

Methane Wavelength Measurement

In a coordinated effort, the wavelength of the 3.39 μ m line of methane was measured with respect to the ⁸⁶Kr 6058 Å primary standard of length by Barger and Hall.⁽²⁾ Using a frequency-controlled Fabry-Perot interferometer with a pointing precision of about 2×10^{-5} orders, a detailed search for systematic offsets inherent in the experiment, including effects due to the asymmetry of the Kr standard line, was made. Offsets due to various experimental effects (such as beam misalignments, mirror curvatures, and phase shifts, phase shift over the exit aperture, diffraction, etc.) were carefully measured and then removed from the data with an uncertainty of about 2 parts in 10^9 . This reproducibility for a single wavelength measurement illustrates the high precision which is available using the frequency-controlled interferometer.

At the fifth session of the Consultative Committee on the Definition of the Meter (CCDM),⁽⁴⁾ results of Barger and Hall as well as measurements made at the International Bureau of Weights and Measures,⁽²⁶⁾ the National Research Council,⁽²⁷⁾ and the National Bureau of Standards at Gaithersburg⁽²⁵⁾ were all combined to give a "recommended" value for the wavelength of the transition of methane used to stabilize the He-Ne laser. The recommended value is

$$\lambda_{\text{CH}_4} [P(7), \text{band } \nu_3] = 3\,392\,231.40 \times 10^{-12} \text{ m} \quad (7)$$

Results

Multiplying this recommended wavelength of methane by the measured frequency yields the following value for the speed of light:

$$c = 299\,792\,458 \text{ m/s} (\Delta c/c = \pm 4 \times 10^{-9}) \quad (8)$$

which was also recommended by the CCDM⁽⁴⁾ to be used in distance measurements where time-of-flight is converted to length and for converting frequency to wavelength and vice versa.

This result is in agreement with the previously accepted value⁽²⁴⁾ of $c = 299\,792\,500 (100) \text{ m/s}$ and is about 100 times more accurate. A recent differential measurement of the speed of light has been made by Bay, Luther, and White⁽²⁸⁾; their value is $299\,792\,462 (18) \text{ m/s}$, which is also in agreement with the presently determined value. A third, recent, highly accurate value was obtained by multiplying the frequency of the R(12), 9.3 μ m line of CO₂ by the wavelength,⁽²⁹⁾ which yields the value of $299\,792\,459.0 (0.8) \text{ m/s}$ for c .

The fractional uncertainty in this value for the speed of light, $\pm 4 \times 10^{-9}$, arises from the interferometric measurements with the incoherent krypton radiation which defines the international meter. This limitation is indicative of the remarkable growth in optical physics in recent years; the present krypton-based length definition was adopted only in 1960.

Various measurements of c since Froome's work are listed in Table 1 and are plotted in Figure 3. One sees a remarkable convergence of the values of c for the first time in history!

REDEFINITION OF THE METER

In addition to the values "recommended" at the fifth CCDM meeting for the wavelength of methane and the speed of light,⁽⁴⁾ the following value for the wavelength of iodine was also "recommended":

$$\lambda_{\text{I}^{127}} R(127), \text{band } 11-5, i \text{ component} \\ = 632\,991.399 \times 10^{-12} \text{ m} \quad (9)$$

These "recommended" values are in agreement with wavelength measurements to the limits possible with the krypton length standard (that is, with $\delta\lambda/\lambda$ about $\pm 4 \times 10^{-9}$). It is "recommended" that stabilized lasers generating either of these two wavelengths be used to make length measurements in the interim before the meter is redefined.

It was also significant that no further work was recommended on the present length standard, the krypton lamp, which is far inferior to a laser and will probably soon be replaced by a laser.

As a result of the recommendations made by CCDM, two different definitions of a new length standard must be considered. First, we can continue as before with separate standards for the second and meter, but with the meter defined as the length equal to

TABLE I
Speed of Light Measurements Since 1958

Year	Author	Ref.	Method	C (km/s)	δC (km/s)
1958	Froome	24	Radio interferometer	299 792.5	0.1
1961	Cutkosky-Thomas	30	Ratio of units	299 791.96	0.8
1965	Kolibayev	31	Electro-optical	299 792.6	0.06
1965	Rank <i>et al.</i>	32	Spectroscopic	299 792.8	0.4
1966	Karolus	33	Electro-optical	299 792.44	0.2
1967	Grosse	34	Electro-optical	299 792.5	0.05
1967	Simkin <i>et al.</i>	35	Radio interferometer	299 792.56	0.11
1971	Bjerhammar	36	Electro-optical	299 792.375	0.060
1972	Bay <i>et al.</i>	28	He-Ne $\lambda\nu$ (0.633 μm)	299 792.462	0.018
1972	Baird <i>et al.</i>	37	CO ₂ $\lambda\nu$ (9 and 10 μm , avg)	299 792.460	0.006
1972	Evenson <i>et al.</i>	3	He-Ne $\lambda\nu$ (3.39 μm) c.g. peak	299 792.4562	0.0011
				299 792.4587	0.0011
1973	Guelachvili	38	Spectroscopic	299 792.46	0.07
1973	Baird, Blaney <i>et al.</i>	37, 39	CO ₂ $\lambda\nu$ (9.32 μm)	299 792.457	0.006
1973	CCDM Recommended	4, 1	He-Ne $\lambda\nu$ (3.39 μm)	299 792.458	0.0012
1974	Blaney <i>et al.</i>	29	CO ₂ $\lambda\nu$ (9.32 μm)	299 792.459	0.0008 ^a

^aDoes not include the uncertainty in the meter standard.

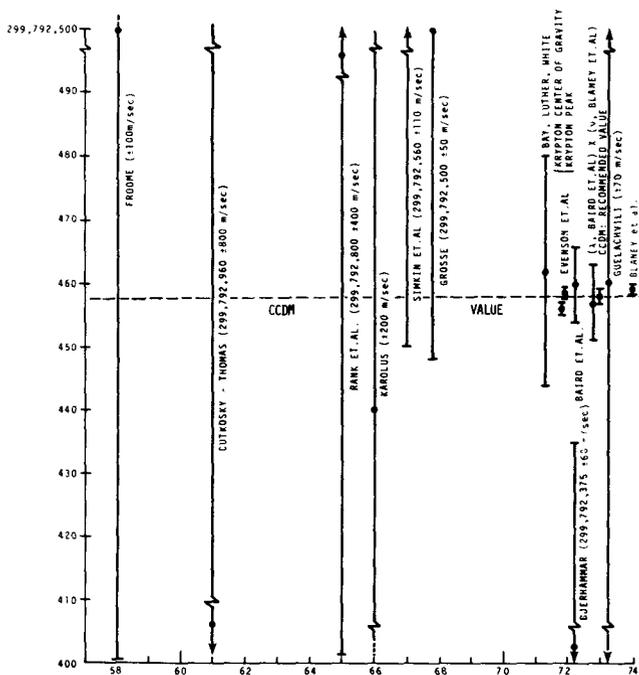


Figure 3. Accurate speed of light values in meters per second since 1958. The value "recommended" by CCDM is 299 792 458 m/s with an uncertainty of about 4 parts in 10⁹. This value maintains continuity in the meter and may be used when the meter is redefined.

$1/\lambda$ wavelengths in vacuum of the radiation from a stabilized laser instead of from a ⁸⁶Kr lamp. Either the methane-stabilized^(6,40) He-Ne laser at 3.39 μm (88 THz) or the I₂-stabilized^(41,10) He-Ne laser at 0.633 μm (474 THz) appear to be suitable candidates. The 3.39 μm laser is already a secondary frequency standard in the infrared, and it is hoped that direct measurements of the frequency of the 0.633 μm radiation will give the latter laser the same status

in the visible. The 3.39 μm laser frequency is presently known to within 6 parts in 10¹⁰, and the reproducibility and long term stability have been demonstrated to be better by more than two orders of magnitude. Hence, frequency measurements with improved apparatus in the next year or two are expected to reduce this uncertainty to a few parts in 10¹¹. A new value of the speed of light with this accuracy would thus be achievable if the standard of length were redefined in terms of the wavelength of this laser.

Alternately, one can consider defining the meter as a specified fraction of the distance light travels in one second in vacuum (that is, one can fix the value of the speed of light). The meter would thus be defined in terms of the second; and, hence, a single unified standard would be used for frequency, time, and length. What at first sounds like a rather radical and new approach to defining the meter is actually nearly one hundred years old. It was first proposed by Lord Kelvin in 1879.⁽⁴²⁾ With this definition, the wavelength of all stabilized lasers would be known to the same accuracy with which their frequencies can be measured. Stabilized lasers would thus provide secondary standards of both frequency and length for laboratory measurements, with the accuracy being limited only by the reproducibility, measurability, and long term stability. It should be noted that an adopted nominal value for the speed of light is already in use for high-accuracy astronomical measurements⁽⁴³⁾; thus, there are currently two different standards of length in existence: one for terrestrial measurement and one for astronomical measurements. A definition which fixes c and unites these two values of c would certainly be desirable from a philosophical point of view.

Independent of which type of definition is chosen, we believe that research on simplified frequency

synthesis chains bridging the microwave-optical gap will be of great interest, as will refined experiments directed toward an understanding of the factors that limit laser optical frequency reproducibility. No matter how such research may turn out, it is clear that ultraprecise physical measurements made in the interim can be preserved through wavelength or frequency comparison with a suitably stabilized laser such as the 3.39 μm methane device.

Frequencies are currently measurable to parts in 10^{13} , and hence the overall error of about 6 parts in 10^{10} for the frequency measurement can be reduced. This measurement was performed fairly quickly to obtain a frequency of better accuracy than the wavelength. It should be possible to obtain considerably more accuracy by using tighter locks on the lasers. For example, the 8 m HCN laser has recently been phase-locked^(44,45) to a multiplied microwave reference, which currently determines the HCN laser linewidths. An improved microwave reference should be a superconducting cavity stabilized oscillator⁽⁴⁶⁾ for best stability in short term (narrowest linewidth) coupled with a primary cesium beam standard for good long term stability.

The relative ease with which these laser harmonic signals were obtained in these frequency measurements indicates that the measurement of the frequencies of visible radiation now appears very near at hand. Even if the point contact metal-on-metal diode is inoperable above 88 THz, conventional nonlinear optical techniques (i.e., second harmonic generation in crystals) could still be used to extend direct frequency measurements to the visible. Such measurements should greatly facilitate one's ability to accurately utilize the visible and infrared portion of the electromagnetic spectrum.

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