Optical Frequency Measurements

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Invited Paper

This paper is a review of the history of the measurement of coherent optical frequencies. Since coherent optical frequency implies a laser device, this is therefore a review of laser frequency measurement. The development of frequency measurement from the Cs frequency standard to the visible is traced. Two related aspects of optical frequency measurements, the speed of light and the redefinition of the meter, are also discussed.

I. Introduction

The absolute frequency measurement of radiation at frequencies above 500 GHz was made possible by the laser: a source of "coherent" radiation [1]. The discovery of continuous-wave (CW) laser oscillation in the infrared regions of the electromagnetic regions in He–Ne gas mixture by Javan *et al.* in 1961 [2] was soon followed by the discovery of CW laser oscillation in the visible [3] and far-infrared regions [4] of the spectrum. The coherence of laser radiation brought about the possibility of the measurement of the frequency of this "coherent" radiation. The first such measurement of a laser frequency (rather than the wavelength) was reported by Hocker *et al.*, in 1967 [5]. The frequencies of single-mode emissions at 890 and 964 GHz (337 and 311 μm) were measured using submillimeter harmonic mixing techniques with silicon-point contact diodes, similar to those developed [6] to generate submillimeter radiation by harmonic generation from millimeter radiation.

These frequency measurements made it possible to use lasers for measuring the speed of light in vacuum c_0. The most reliable measurement of the speed of light at that time was considered to be that made by Froome in 1958 [7]

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in which the frequency f and the vacuum wavelength λ_0 were measured for stabilized microwave radiation in the 4-mm band. The result was given by $c_0 = f\lambda_0$. The main source of uncertainty in this experiment was in the wavelength determination, due to diffraction in the interferometer apertures and from the correction for the refractive index of the air. Froome's one-sigma uncertainty was ± 3 parts in 10^7, but there was disagreement with other results, particularly long-baseline ones (Bergstrand) [8]. The speed of light is used for electromagnetic distance measurement, especially in critical applications such as geodetic-survey baseline measurements, which were at that time limited by the uncertainty of c.

It was apparent by the mid-1960s that a more accurate determination of λ could be made at the shorter wavelength afforded by laser radiation; hence a more accurate value of c could be obtained. This possibility enabled the development of laser frequency measurement programs at various national standards laboratories such as the National Bureau of Standards at Boulder, CO, USA, the National Physical Laboratory, Teddington, UK, and the National Research Council, Ottawa, Canada.

Important steps in the development of laser frequency measurements were the introduction of the metal–insulation–metal (MIM) diode and the demonstration of high-order mixing (12th harmonic of the HCN laser against an H_2O laser at 28 μm) by Evenson *et al.* [9]. Cryogenic Josephson junctions were proving useful for very-high-order mixing with submillimeter lasers [10], [11]. In 1972 accurate frequency measurements were extended to 3.39 μm [12], with a chain of five lasers using MIM diode harmonic mixing devices to generate the laser harmonics.

For accurate frequency measurements it was necessary to have sufficiently stable "targets" to measure. These were provided by narrow atomic or molecular absorption or emission features used to stabilize ("lock") the frequency of the laser. Narrow references were provided by saturated-absorption features observed in passive gas cells at low pressure. These were free of the first-order Doppler effect. Particularly important were the methane-stabilized He–Ne laser at 3.39 μm [13] and the CO_2 laser locked to a saturated fluorescence absorption in CO_2 itself [14]. The methane-stabilized laser was notable for its reproducibility ($\pm$ a few parts in 10^{11}) and compactness, and the stabilized CO_2 laser was a very useful device because of the large number of closely spaced emissions that could be accurately characterized. The I_2-stabilized He–Ne laser oscillating at 633 nm (red) [15] was used as a wavelength standard for laboratory length measurement; but the measurement of its higher optical frequency was foreseen as a much more challenging problem.

"Optical" frequency measurements, therefore, consist of the measurements of these narrow spectral features used to stabilize the lasers, and the metrological landmarks consist of accurate measurements of these features. Most of the early measurements were aimed at determining the speed of light. The first of these highly accurate determinations of the speed of light was made in 1972 at the National Bureau of Standards in Boulder by measuring the frequency and wavelength of the 88-THz (3.39-μm) methane transition. A value for the speed of light (299 792 458 m/s) was recommended the following year by the Consulative Committee for the Definition of the Meter (CCDM) [16]. This value was eventually to become the "fixed" value c_0 used in the redefinition of the meter. It originally had an uncertainty of ± 4 parts in 10^9 which arose almost entirely from difficulty in comparing wavelengths with the ^{86}Kr emission used as the length standard. The recommended value was based on the NBS Boulder speed of light measurement together with values of the wavelength from three other laboratories. The CO_2 frequency measurement used to measure methane was also confirmed by an independent measurement at NPL, Teddington [17].

The 1973 recommendation for c_0 was made to meet the requirements of astronomers using lasers to measure astronomical distances. They used the speed of light to convert electromagnetic wave transit times into distance. (The time to the moon and back was being measured to about 1 part in 10^{10}.) Various speed of light determinations then in progress were completed and the methane frequency at 88 THz was directly checked by a measurement at NPL [18]. There was excellent agreement in all these measurements.

After the initial speed of light determinations, work on optical frequency measurement followed three broad directions. These were first to extend frequency measurement to higher frequencies than 88 THz, particularly towards the visible region, in the hope of unifying the standards of time and length. Second, the more accurate stabilized lasers were of interest as potential frequency standards and it is desirable to know their frequencies to accuracies close to their reproducibilities. A third direction was the use of laser frequency measurements for ultra-high-resolution, high-accuracy spectroscopy. The technique is extremely important in understanding complicated molecular spectra such as those from SF_6 and OsO_4 [19]–[21], and SiF_4 [22].

The extension of optical frequency measurements to 260 THz (1.15 μm) was performed at NBS Boulder through a sequence of gas lasers at 2.03, 1.52, and 1.15 μm [23]. Harmonic mixing was not obtained in the MIM diode above 200 THz (1.5 μm) consequently, phase-matched second-order effects in nonlinear crystals were used for frequency addition or doubling above 200 THz. An I_2 transition at 520 THz (576 nm) in the yellow–green region was shown to be an excellent stabilization reference for frequency-doubled light from the 1.5-μm He–Ne laser at the National Research Council (NRC), Ottawa [24]. This was followed by a joint NBS–NRC experiment in 1979 which measured the frequency of this transition to provide the first frequency measurement of a visible radiation [25].

A series of measurements of the methane transition used to stabilize the He–Ne laser at 88 THz was achieved between 1978 and 1981 at NPL [26], the Laboratoire Pour Temps et Fréquences (LPTF) [27] Paris, France, and in two laboratories in the USSR [28]–[30]. The methane-stabilized laser was used as a standard at NBS Boulder in making a more accurate measurement of the visible I_2 transition at 520 THz (576 nm) and a red I_2-stabilized He–Ne laser at 474 THz (634.3 nm) [31], [32]. The uncertainty in these measurements was ($\pm 1.6 \times 10^{-10}$), and provided frequency values 10 times more accurate than the corresponding wavelength could be determined from the Krypton-86 length standard. The latter experiment made use of excited Ne atoms for resonant three-frequency addition of 88, 125, and 260 THz (3.39, 2.39, and 1.15 μm, respectively) [33]. The NBS experiments used for the first time a tunable color-center laser as a transfer oscillator, to provide a source at half the frequency of the 1.15-μm He–Ne laser (2.3 μm). This same laser was also used instead of a 2.39-μm He–Ne laser to provide sufficient power for making the 633-nm measurement.

The accurate frequency measurements permitted the adoption of a new definition of the meter by the General Conference of Weights and Measures in Paris in 1983 [34]. The new definition adopted an exact fixed value for the speed of light: the value recommended in 1973. The "Mise en pratique," recommends a list of stabilized-laser frequencies to be used to realize the meter [35]. The most accurate values in the "Mise en pratique" from the visible region have 1-sigma uncertainties of 4 parts in 10^{10}, over an order of magnitude better than the 4 parts in 10^9 uncertainty of the previous length standard, the ^{86}Kr lamp. The "Mise en pratique" can be revised whenever better measurements become available.

The following text summarizes some of the important techniques of laser frequency measurement, describes some illustrative frequency-synthesis chains, lists accurate measurements of the speed of light, and discusses laser frequency standards. Recent proposals for more efficient harmonic generation and for laser-based frequency standards are also discussed. No replacement of the cesium frequency standard is imminent, but a methane-stabilized laser "clock" [36] has been demonstrated; it provides a less accurate, but interesting alternative to a cesium clock at a much higher frequency.

II. Techniques of Optical Frequency Measurement

A. Nonlinear Devices for Harmonic Generation and Mixing

Up to about 150 THz (2 μm) almost all frequency measurements have been made using essentially wide-band nonlinear point-contact devices, notably the MIM, the metal to semiconductor, Josephson-junction, and Schottky diodes. This is a perhaps surprising extension of the range of the early RF and microwave cat-whisker diode. Above about 150 THz, bulk nonlinear dielectric crystals have been used for second-order effects, frequency doubling, and addition or subtraction of frequencies. In one experiment, excited Ne atoms in the gaseous state served for the summation of three frequencies [32], [33]. Mixing (difference–frequency generation) above 150 THz has been achieved with photodiodes, although most commercial diodes are limited to difference frequencies substantially less than 100 GHz.

Reviews concentrating on nonlinear devices for laser frequency measurement have been given by Knight and Woods [37] and by Klementev *et al.* [38]. Thus opportunities for frequency synthesis are, at present, more restricted above 150 THz than below.

Information on the highest harmonic order, highest frequency reached, and the largest beat frequency observed in the photo-mixing regime is given for the principal kinds of mixing device in Table 1.

Most of the devices used operate through a resistive rather than reactive nonlinearity, and none of these is highly efficient for harmonic generation. Each has a useable frequency range determined mainly by the physical characteristics of the devices, and in practice, this is more important than ease of harmonic generation. Thus Josephson junctions generate high-order harmonics easily, to mix with frequencies up to 1 or 2 THz but become much less effective at 4 THz. For niobium, the most practical superconductor so far, the paired-electron band-gap energy corresponds to a frequency of 0.8 THz. The MIM diode by comparison with a semiconductor diode has only about 1/100th of the spreading resistance in the post material, thus offering a 100-times higher cutoff frequency, even though the voltage–current characteristic is markedly less nonlinear than that of most semiconductor diodes. The GaAs Schottky diode is an improvement on the silicon point-contact diode. It exhibits a higher mobility of the semiconductor, and a rugged and highly "ideal" voltage–current characteristic of the barrier formed with the evaporated metal contact. Both MIM and Schottky diodes have recently proved useful at visible-laser frequencies for obtaining large difference frequency signals [47], [48]. Mixing in phase-matched bulk crystals all generate much larger difference frequencies but there are problems in phase-matchability and transparency of the available crystals. The individual crystal also has a limited tuning range, obtained by varying its temperature or the beam angles.

Since the main characteristics of MIM diodes have been

Table 1

	Wide-Band Action—Highest Harmonic			Harmonic Mixing Highest Frequency			Second-Order Action High-Frequency Beat		
Device	n_{MAX}	f/THz	Ref.	f/THz	n	Ref	Δf_{MAX}/GHz	f_0/THz	Ref
Semiconductor PC diode (W–Si)	23	1.58	[39]	3.56	4	[42]			
Josephson junction PC	825	0.89	[11]	4.25	43	[43]	109	30	[46]
(Nb–Nb)	401	3.8	[10]						
Schottky diode (GaAs)	33	2.5	[40]	5.3	13	[44]	900	528	[47]
Metal–Insulator–Metal (MIM) diode	12	10.7	[9]	197	$f_1 + f_2 \sim 2$	[45]	2500	~ 500	[48]
(W–Ni)	16	26	[41]	130	$3f_1 + 2f_2 \sim 5$	[31]			
Avalanche photodiode (GaAs)							100	~ 500	[49]
Three-terminal FET (GaAs)							300	474	[50]

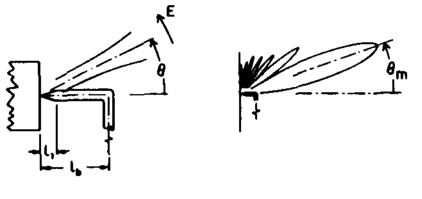

Fig. 1. The MIM diode optical antenna. (a) Showing both the conical antenna part ℓ_τ and the long wire antenna part ℓ_b. (b) The calculated antenna pattern for $\ell_\tau = 7\lambda$.

published [51]–[53] brief notes will follow on investigations relevant in an engineering sense.

The usual room-temperature mixer, a MIM or a Schottky diode, has an open structure (without waveguide enclosures) and radiation is coupled to it with lenses or mirrors. The whisker is usually in the horizontal plane so that the face of the post is vertical. The whisker acts as a long antenna for laser radiation up to about 30 THz (Fig. 1), having a length of several wavelengths, and a right-angle bend serves to define the end of the antenna. Above about 30 THz, coupling to the diode occurs at the conical end of the antenna. The cone is made by electrochemically etching the tungsten, and this conical antenna then produces the electrical currents at the tip.

The long antenna has a series of lobes, the first and largest having a maximum at an angle θ_m to the antenna, given by $\theta_m = \cos^{-1}(1 - 0.371\lambda/L)$, where L is the effective antenna length. The lobes are axially symmetrical about the whisker. The conical antenna exhibits a broader, simpler, but somewhat similar pattern.

In practice, the laser radiation is focussed from a Gaussian beam onto the antenna and the angle is adjusted to the peak of the first lobe. With the whisker horizontal, all laser radiation applied is horizontally polarized and each wavelength is arranged at an angle near its own θ_m in the horizontal plane. To achieve optimum coupling, the focal length and beam diameter should be chosen to match the converging beam to the half-width of the antenna lobe. Corner reflectors have also been used to improve coupling to the diode [54].

Frequencies from dc up to perhaps 10 GHz can be coupled in or out of the diode directly by coaxial lines or with short antennas on the coax. Care is necessary to isolate the extremely delicate point contact (especially in the case of MIM diodes) from mechanical forces on the coaxial lead. Millimeter-wave frequencies, typically 20–100 GHz, are applied to the whisker from an open waveguide, placed above the diode, with the E-vector parallel to the whisker.

In some situations there may be three laser beams and microwave radiation on the diode, and the heterodyne frequency of the order of 10–600 MHz extracted. It is not easy for all radiations to be optimally coupled at the same time. Usually, higher power and better coupling are required for the radiation having the highest harmonic. Excessive power from the radiation at the fundamental frequency generally increases the diode noise and diminishes the signal obtained. With laser radiation applied to MIM diodes, increases in noise of the order of 10 dB are seen at low frequencies, decaying into the white noise at frequencies above about 30 MHz. Laser powers of the order of a microwatt (in the FIR region) produces an excellent signal-to-noise ratio in low-order mixing experiments on the MIM diode; higher powers can be beneficial up to about 200 mW in the IR region, at which point destructive whisker heating takes place.

B. Laser Frequency Measurement and Frequency-Synthesis Chains

In general, when various frequencies of electromagnetic radiation are incident on a nonlinear detector-mixer, such as the MIM diode, Schottky diode, or the Josephson junction, all harmonics and sums and differences are generated. A low-frequency difference, Δf (usually between 30 and 1500 MHz), is used to measure unknown frequencies as will be shown in the following discussion.

1) Mixing Microwave Harmonics with a Laser: The simplest scheme for measuring a laser frequency f_L is by beating directly against a harmonic of a known microwave frequency f_M using a mixer. The value of f_L is calculated from measuring the beat frequency Δf, its sign, and the harmonic order n, in the expression

$$f_L = nf_M \pm \Delta f. \tag{1}$$

f_L is the frequency of a laser which, in the case of a gas laser, is tunable by about ± 2 parts per million (ppm). The measurement accuracy can exceed 1 part in 10^{11}. However, such reproducibility and accuracy are achieved only when a laser stabilized to sub-Doppler atomic or molecular absorption is being used. Both n and the sign of Δf can be established by varying f_M, and for this purpose a wide-band spectrum analyzer of order 1 GHz is useful.

The microwave oscillator (usually a klystron oscillating at less than 100 GHz) can be phase-locked to a quartz crystal. In this case, the crystal phase noise is multiplied right up to the laser frequency and the beat signal is broadened, thus the resolution of Δf is reduced [55].

Alternatively, the microwave oscillator can be phase-locked to the laser being measured. In this case, the phase-lock signal originates from the phase angle between the heterodyne signal Δf and a crystal-controlled IF reference. Quartz-crystal frequency multiplication is not then required beyond the microwave frequency, although signals have been shown to be clean enough (16-dB signal/noise in 100-kHz bandwidth) from a simple 120-MHz quartz-crystal oscillator when multiplied to 2.5 THz [56]. The phase-lock loop (PLL) bandwidth must be wide enough to suppress the klystron-produced noise at the laser frequency. In a simplified methane-frequency chain, a 99-GHz reflex klystron has been phase-locked to a laser at 4.25 THz (70 μm) by 43rd-harmonic mixing in a Joseph junction using a servo-loop with an IF bandwidth of 6 MHz and an overall natural frequency near 300 kHz [26], [46].

2) Two-Laser Mixing: The lasers used are usually gas lasers whose frequencies are selected from those available. A microwave frequency f_M is often needed to produce a final beat frequency Δf in the RF region. Thus the two-laser harmonic-mixing experiment is of the form

$$f_{L2} = n_1 f_{L1} \pm f_M \pm \Delta f \tag{2}$$

where f_{L2} is the unknown frequency and f_{L1} is the known one.

In such a measurement f_M can be phase-locked to a quartz-crystal oscillator. It is sometimes useful to phase-lock f_{L1} to f_{L2}, or to lock f_M to the laser pair via Δf. In order to obtain a fast means of controlling a laser frequency for phase locking, a long-range relatively slow PZT mirror controller is used in conjunction with a high-speed device such as a discharge-current controller [57], a small PZT [58], Stark cell [59], or intra-cavity electrooptic crystal [60], [61]. Fast frequency control by extracavity devices has also been demonstrated [62].

3) Three-Laser Mixing: When two suitable lasers do not exist for two-laser mixing, a third laser may be introduced according to the scheme

$$f_{L3} = n_1 f_{L1} \pm n_2 f_{L2} \pm f_M \pm \Delta f \qquad (3)$$

where f_{L3} is the unknown frequency and f_{L1}, f_{L2}, and f_M are known. This has recently been used to measure a color-center laser's frequency with two stabilized CO_2 lasers [31].

Three lasers are also used when one laser's frequency corresponds to the difference between the frequencies of two lasers needing to be compared. Then n_1 and n_2 are one and minus one, respectively. This has recently been demonstrated with MIM mixers for spectrally narrow visible dye lasers separated by 2.5 THz [51]. The "high-frequency beat" section of Table 1 shows the achievements with different mixers for this kind of mixing.

4) Laser Frequency-Synthesis Chains Involving Several Stages of Mixing: To attain the highest frequencies in the processes just described, a sequence of frequency measurements are combined in a so-called chain of lasers. The construction of these laser frequency-synthesis chains depends upon a combination of the available lasers and the obtainable properties of the nonlinear mixing elements summarized in Table 1. CW gas lasers have mainly been used because of their simplicity and spectral purity. Power output deficiencies have been overcome by building gain tubes up to 8 m long [9]. The set of several hundred CO_2 frequencies (from seven isotopic species) [63], covering the spectrum from 25 to 30 THz (9 to 11 μm), is widely used in synthesis from the microwave region to the 100–150-THz (3–2-μm) region.

Above 100–150 THz, the average spacing between gas-laser frequencies increases and the bandwidths of available frequency-mixing devices become smaller. Thus the CW tunable-laser systems have a role in simplifying frequency-synthesis schemes, in spite of their added complexity. An important example is the use of a color-center laser at 2.3 μm by Pollock *et al.* [31] to increase the measurement accuracy of the previously mentioned chain to 520 THz [32].

In the submillimeter region it is a fortuitous accident that three discharge lasers: the HCN laser at 337 μm, the D_2O laser at 84 μm, and the H_2O laser at 28 μm, produced convenient harmonically related frequencies connecting to a CO_2-laser line at 32 THz (9.3 μm). Only a few lines were available from these discharge lasers; however, this soon changed with the discovery of optically pumped far-infrared (FIR) lasers (pumped by CO_2 laser radiation) in 1970 [64]. By 1980 there were 1200 such CW emissions [65], [66] and by now there have been perhaps 1000 more reported. It is important to note, however, that comparatively few of these are powerful (10 mW or more) and that again comparatively few emit at frequencies above 3 THz (100 μm). Thus some current synthesis chains use three-laser mixing to access specific CO_2 frequencies [67], [68]. Also a chain at NRC, Ottawa by Whitford [69], [70] uses only CO_2 lasers and MIM diodes and utilized the difference frequencies between CO_2 lasers as transfer oscillators to multiply from the microwave region.

Some examples of frequency-measurement chains are shown in the following figures. The first chain to methane in 1973 [12] is shown in Fig. 2. Note the use of the HCN

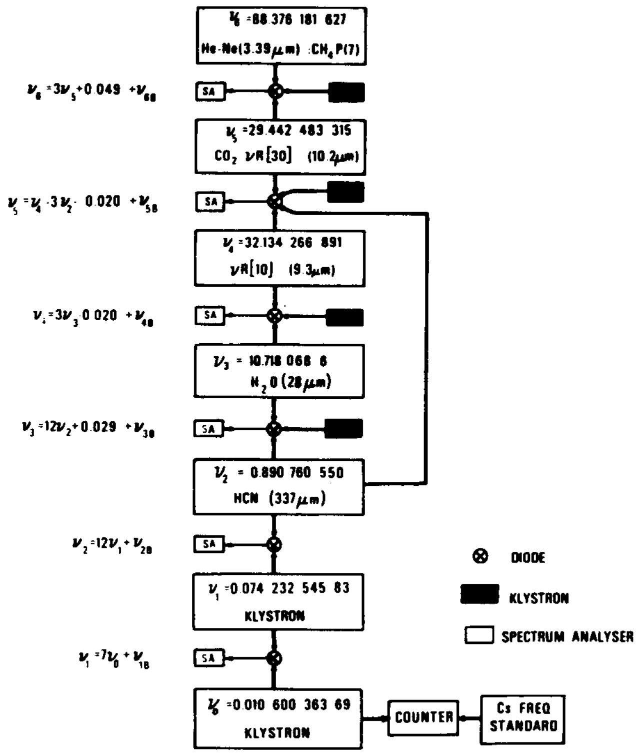

Fig. 2. The first laser frequency synthesis chain to frequency measure the He–Ne CH_4-stabilized laser. (All frequencies in THz.)

laser twice, in the chain to the H_2O laser, and separately to measure the difference frequency between the CO_2 lasers. Stabilized-CO_2 lasers were used as transfer standards in the chain. For more accurate measurement of the methane frequency the chain shown at Fig. 3 was used [26]. This chain used laser transfer oscillators at only two intervening frequencies and used simultaneous counting of beat frequencies to eliminate the effect of their frequency fluctuations. The frequency mixing range of the Josephson junction was extended for this experiment to 4.25 THz.

A third accurate chain to methane [30] is shown at Fig. 4. Here the various oscillators were phase-locked together and counting was performed at only one point in the chain: at 29.5 THz.

The extension of accurate measurement to a visible frequency, already mentioned, was performed in 1983 [31] using the scheme in Fig. 5. The target was an I_2 transition at twice the frequency of the 1.15-μm (260-THz) He–Ne line. A methane-stabilized laser was used to measure the frequencies of the two well-controlled stabilized CO_2 lasers

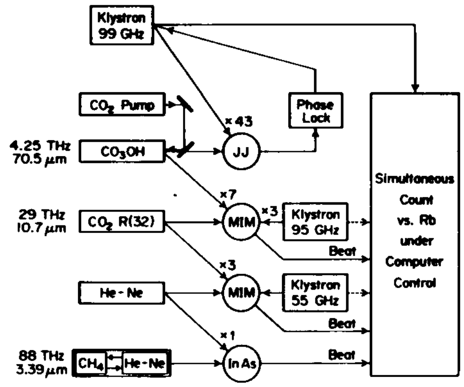

Fig. 3. A simultaneous-counting scheme to frequency measure the He–Ne CH_4-stabilized laser.

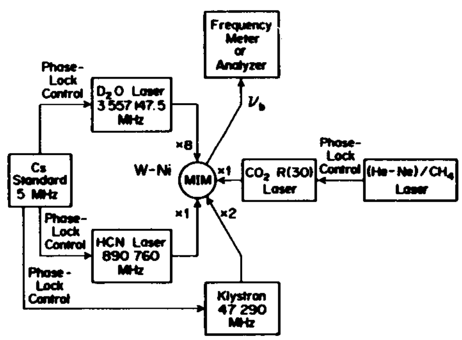

Fig. 4. A phase-locked method for the He–Ne CH_4-stabilized frequency measurement.

used at the bottom of the chain. This scheme is more complicated than that for methane in Fig. 3 in spite of its smaller frequency-multiplication range, because of the limitation to frequency-doubling stages above 130 THz (2.3 μm).

The scheme previously mentioned that made use of the energy levels of the Ne atom to sum the 3.39-, 2.39-, and 1.15-μm emission of the He–Ne laser [32] is shown in Figs. 6 and 7. Radiation at exactly the sum of the three radiations traversing the tube is emitted and corresponds to the well-known 633-nm red emission. It is, however, a sum frequency due to resonant four-wave mixing in Ne, and is not a laser emission [33]. This scheme was used by the group at NBS, Boulder to transfer from their frequency measurement of an I_2 transition at 576 nm to the I_2-stabilized He–Ne laser at 633 nm. The 1.15-μm laser was doubled and stabilized to the previously measured I_2 transition at 576 nm; the color-center laser was used instead of He–Ne at 2.39 μm, and was measured with respect to methane as is shown in Fig. 5; and the 3.39-μm laser was directly compared with methane. Thus the 633-nm emission produced was known in terms of the methane frequency. The frequency of the iodine line used to stabilize a He–Ne laser was measured by measuring the RF beat produced by the two beams in a photodiode.

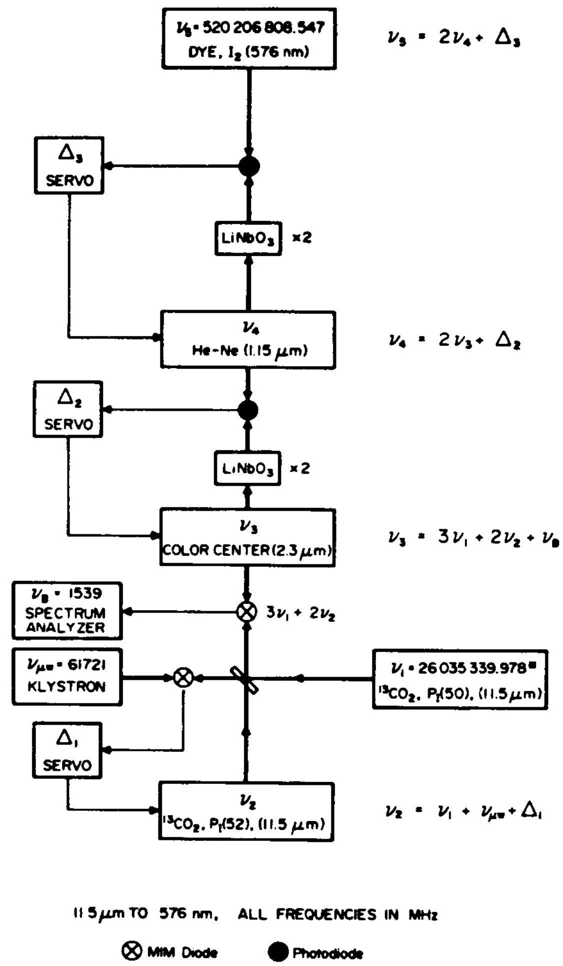

Fig. 5. The first laser frequency chain to the visible. The target was a molecular iodine transition at 520 THz.

III. Results

A. Measurements of the Speed of Light

A summary of values for the speed of light obtained with lasers is shown in Fig. 8. Apart from the first experiment listed [71], which was notable for the economy of apparatus, all others involved the measurement by harmonic generation and mixing techniques of the frequency of a stabilized laser. The laser's wavelength was separately measured by comparing its wavelength with the emission from a ^{86}Kr lamp by which the meter was realized either by direct interferometry [88] or by a technique involving upconversion [73], [75], [98]. It was recognized in 1973 that the main uncertainty arose from the imprecision of measurements based on the ^{86}Kr lamp so that the CCDM recommended value was accompanied by the expression that it would be desirable to keep this value of c_0 for a possible future redefinition of the meter. A new definition of the meter was adopted in 1983 by retaining and fixing this value, after the intervening measurements (Fig. 8) showed i) that there was no serious discrepancy in the value adopted in 1973 and ii) that the frequency values of the visible laser radiations in common use as laboratory length standards had been established more accurately than their corresponding wavelength values.

B. Absolute Frequency Measurements of Stabilized Lasers

Early frequency measurements of metrological importance were those on the CO_2 fluorescence-stabilized CO_2 laser [90], [91]. These were used to calibrate accurate interline measurements of CO_2 and the associated determination of its molecular constants. The CO_2 frequencies then

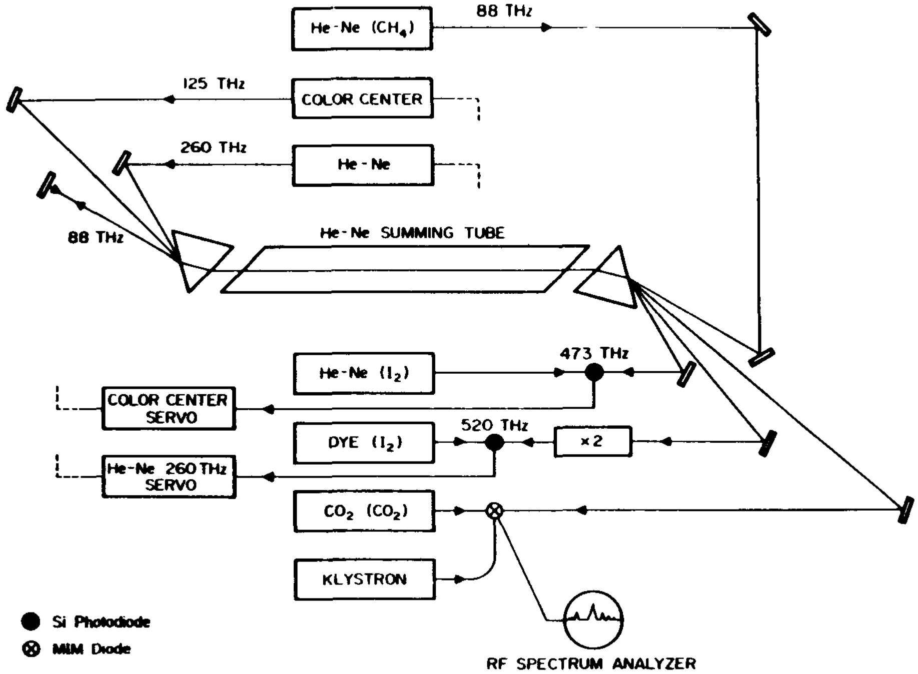

Fig. 6. A schematic diagram of the method used to frequency measure the molecular iodine transition used to stabilized the visible He–Ne laser at 473.6 THz (633 nm).

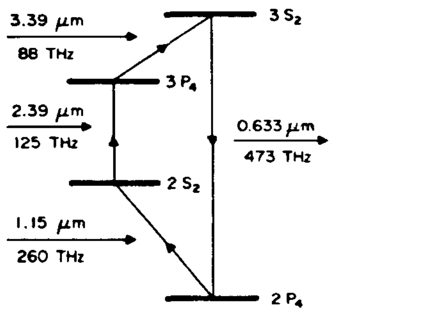

Fig. 7. A partial energy level diagram of Ne showing the energy levels involved in summing to obtain output at 473.6 THz.

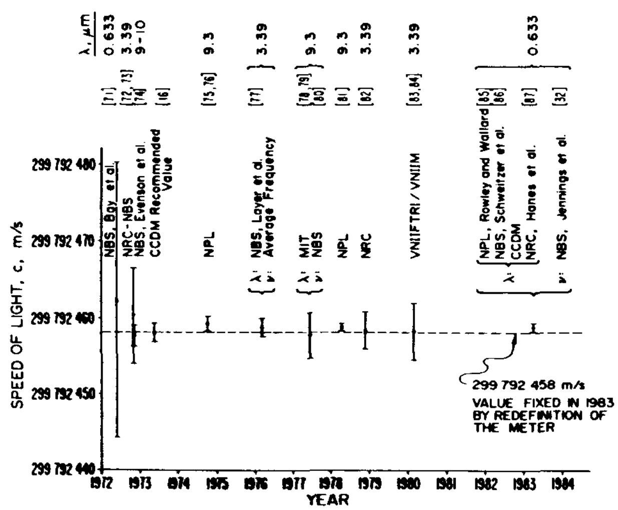

Fig. 8. Laser speed of light measurements from 1972 to 1984.

obtained provided a grid of frequency references between 25 and 33 THz (9 to 11 μm) and were accurate to about 1 part in 10^9.

Fluorescence-stabilized CO_2 lasers were used as transfer oscillators to obtain measurements of the frequency of the He–Ne laser locked to the $F_2^{(2)}$ transition of methane at 88 THz (3.39 μm). These measurements [12], [18] were accurate to about 5 parts in 10^{10}, and agreed within 2 parts in 10^{10}.

Numerous measurements have been made on gas lasers in the submillimeter region in more than a dozen laboratories around the world. About one third of the 2000 or more emissions known in 1984 had been characterized by frequency measurement. These measurements mostly refer to the center of the laser tuning profile and are, therefore, comparatively less accurate, about 2 parts in 10^7. However, they are better than any wavelength measurements and are of considerable value for far-infrared spectroscopy.

A list of the lines ordered by frequency is available for submillimeter laser transitions with wavelengths larger than 12 μm [65], and a list of frequency measurements of optically pumped far-infrared laser lines is in preparation [66].

After the first CO_2 and methane frequency measurements, the speed of light was satisfactorily determined, and several laboratories made more accurate measurements of the frequency of the methane transition used to stabilize the 3.39-μm He–Ne laser. From the beginning, this laser had shown promise of accuracy close to 1 part in 10^{11} [13], but later attempts to improve this pointed to an estimated limit of about 1 part in 10^{13} [92]. Nonetheless, this is so far the most accurate stabilized laser in current use. Eight measurements from four laboratories have been reported since 1979 at an accuracy better than about 1 part in 10^{10}. A comparison of the various measurements since 1973 is shown in Table 2.

The Soviet Laboratories, VNIIFTRI, Moscow [29], [93] and the Institute of Thermophysics, Novosibirsk [28], followed

Table 2 Frequency Measurements of the $F_2^{(2)}$ Methane-Stabilized He–Ne Laser at 88 THz (3.39 μm)

Date Published Day	Mo.	Yr	Reference	Institution	ν − 88 376 181 000 kHz	σ kHz
11	Nov.	72	[72]	NBS	627	50
2	Feb.	73	[12]			
17	Apr.	75	[18]	NPL	608	43
5	Sept.	79	[29]	VNIIFTRI (GS)	586	10
12	Dec.	80	[93]			
	Dec.	79	[82]	NRC	570	200
12	Dec.	80	[27]	LPTF	618	14
12	Dec.	80	[26]	NPL	616	3
	Dec.	81	[94]	ITP	603	3.0
	May–June	83	[36]			
12	Dec.	81	[35]	LPTF	612	11
20	Aug.	81	[30]	VNIIFTRI (GS)	603.4	1.4
	Oct.	83	[35]	[Recommended value	608	3.9]
11	Nov.	83	[95]	ITP	602.9	1.2
	Jan.	85	[96]			
6	Jun.	85	[67]	LPTF	600.0	3.4

broadly the original scheme of Fig. 2 but improved the accuracy by extensive use of phase locking. This was carried so far in Novosibirsk as to provide the downward phase-locking methane clock previously mentioned [36]. This clock is now said to operate typically for 10–20 min without interruption. This is the only methane $F_2^{(2)}$ frequency measurement to have been made with the hyperfine structure resolved. The second measurement at VNIIFTRI [30] was made with the scheme of Fig. 4.

The measurement at LPTF in France [27] also broadly followed the scheme of Fig. 2, and exhibited improved laser stability, and phase-locked operation of the lowest frequency laser. A measurement at NPL [26] achieved improvement in resolution by using the much-simplified synthesis scheme in Fig. 3, and by simultaneously counting the beat frequencies (for 1 s) to eliminate the instability effects of the transfer lasers. A recent measurement at LPTF [67] makes use of fully phase-locked counting of an OsO_4-stabilized laser at 10.3 μm with $\pm$50-Hz resolution.

The accuracy of these experiments demonstrates that the techniques of laser frequency measurement are capable of approaching the reproducibilities (accuracies) of the stabilized lasers being measured. This accuracy compares with that of the available rubidium standards and approaches that of even the cesium standard used.

The advances made at NBS Boulder in the extension of frequency measurement beyond 88 THz, to 260 THz (1.15 μm) and to the visible region of the spectrum, are shown in Figs. 5 and 6 [31], [32]. The scheme involved frequency multiplication from stabilized CO_2 lasers. These were calibrated against a methane-stabilized laser at 88 THz (the JILA telescope laser), assuming, from foregoing measurements (Table 2), that

$$f_{CH_4} = 88\,376\,181.609 \pm 0.009 \text{ MHz}.$$

The frequency of the ^{20}Ne Lamb-dip stabilized He–Ne laser at 1.15 μm was found to be

$$f_{1.15} = 260\,103\,249.26 \text{ MHz}$$

with a total fractional one-sigma uncertainty of 3.1 parts in 10^{10}. The frequency of the "o" hyperfine component of the $^{127}I_2$ 17-1 P(62) transition at 576 nm was found to be

$$f_{576\text{“o”}} = 520\,206\,808.547 \text{ MHz}$$

with a total fractional one-sigma uncertainty of 1.6 parts in 10^{10}.

The "o" transition of I_2 thus measured was used to measure the "i" and "g" hyperfine components of I_2 at 633 nm by the mixing process portrayed in Figs. 6 and 7 [32]. The frequency was likewise referred to the methane-stabilized laser assuming the value just given. The results for the $^{127}I_2$ 11-5 R(127) transition was

$$f_{633\text{“i”}} = 473\,612\,214.830 \pm 0.074 \text{ MHz}$$

where the I_2-stabilized laser was adjusted to the CIPM-recommended operating conditions [35].

These measurements in the visible part of the spectrum have accuracies of $\pm$1.6 parts in 10^{10} which is within a factor of three of the best accuracy of the stabilized lasers used in the lower part of the chain.

A list of recommendations of the best known values for the frequencies of stabilized lasers of interest as length standards was prepared by the Consultative Committee for the Definition of the Meter in 1983 [35], and is shown in Table 3. The list accompanies the new definition of the meter.

The list takes account of the preceding frequency mea-

Table 3 CIPM Recommended Stabilized Laser Frequencies [35]

Laser	λ(nm)	Stabilization Ref.				ν(THz)	$\Delta\nu/\nu$ (3-sigma)
He–Ne	3392	CH_4	ν_3	P(7)	$F_2^{(2)}$	88. 376 181 608	$\pm 1.3 \times 10^{-10}$
Dye	576	$^{127}I_2$	17-1	P(62)	0	520. 206 808 51	$\pm 6 \times 10^{-10}$
He–Ne	633	$^{127}I_2$	11-5	R(127)	i	473. 612 214 8	$\pm 1 \times 10^{-9}$
He–Ne	612	$^{127}I_2$	9-2	R(47)	0	489. 880 355 1	$\pm 1.1 \times 10^{-9}$
Ar^+	515	$^{127}I_2$	43-0	P(13)	a_3	582. 490 603 6	$\pm 1.3 \times 10^{-9}$

surements discussed above and will be seen to assign a greater uncertainty than those of individual frequency measurements. The reasons are that i) a three-sigma uncertainty is given, rather than the usual one-sigma value, ii) the measurements of methane disagreed by about three times one-sigma, and iii) for some visible frequencies additional data were taken into account.

The uncertainties in Table 3 are about an order of magnitude larger than the accuracies claimed for the lasers themselves, so that improved measurements can be expected in the future.

Schemes to measure the frequencies of stabilized lasers in the visible spectral region are being explored at various laboratories. A suitable stabilized laser at 576 nm has been evaluated at NPL [97] and a frequency synthesis experiment has been described [68].

A summary of stabilized-laser frequency measurements traceable to cesium, and with accuracies exceeding 3 parts in 10^9, has been compiled [98]. Recent measurements of high accuracy have been made on an OsO_4 transition near 29 THz (10.3 μm) [67] and on the methane E line at 88.373 THz (3.39 μm) [95], [96].

IV. Discussion

A. Sources of Error in Laser Frequency Measurement

1) Inexactitude in Frequency-Mixing Devices: When the nonlinear properties of systems with well-defined energy levels are being used for frequency mixing it is important to guard against quanta characteristic of the mixing device (as opposed to those of the input frequencies) contributing to the output frequency. Checks can be made, by varying the input frequencies, for example.

On devices used for harmonic generation, some experimental checks have been made for differences of this type. An indirect check on the agreement between Josephson-junction harmonic mixing and the use of entirely room-temperature harmonic mixers is afforded by the results for the 88-THz methane frequency in Table 2. The measurements from NPL alone used the Josephson junction. In a specific test at NPL, see [76, pt. I, Appendix], frequency multiplication to 0.89 THz in a Josephson junction was found to agree with that in an MIM diode within a 1-sigma uncertainty of ± 1.2 parts in 10^{10}. (The test was primarily to compare frequency measurement by frequency division and counting with that by observation of a beat on a spectrum analyzer, but the harmonic mixers were also different.)

A comparison of frequency doubling in a bulk nonlinear crystal and in an MIN diode has been made by Edwards and colleagues at NPL [61]. A CO_2 laser's emission was doubled in a crystal of CdGeAs, and was also doubled in an MIM diode. Then the two frequencies were compared. Any net difference arising in the two frequency-doubling processes was found to be less than 7 parts in 10^{16}.

2) Effect of Phase Noise in Sources Used for Frequency Multiplication: The problem of spectral broadening in harmonics as a result of phase noise in the fundamental-signal oscillator showed at an early stage in laser frequency measurement. Lasers provided clean local oscillators with which to observe harmonic signals multiplied from quartz crystals, for example, and from microwave oscillators.

In the beginning, phase noise from quartz crystals used for phase-locking klystrons was a problem, but by using simplified high-frequency quartz-crystal oscillators (near 100 MHz) and straight frequency multipliers (avoiding some kinds of synthesizers) clean signals could be observed to 2.5 THz [56].

Studies on the special requirements of laser frequency measurement and on the properties of multiplied quartz-crystal noise [55] have helped to clarify the problem, specifically as to the point of catastrophic spectral broadening.

The use of downward phase-lock loops has diminished demands on upward frequency multiplication. High loop bandwidths of order 300 kHz have permitted downward phase locking a 99.6-GHz reflex klystron to a 4.25-THz laser via 43rd-harmonic mixing in a Josephson junction [46].

3) Doppler Effect and Time-Dependent Phase Shifts: At high accuracies it is necessary to guard against the effects of relative motion of different parts of the apparatus which might not cancel for the different methods used to observe signals, such as stored spectrum analyzer displays. Another potential source of error is from phase shifts in signal paths or filters which drift monotonically with time, as from a slowly changing temperature. Discrepancies at the level of 1 part $\sim 10^{12}$ have been observed [26, Appendix 1].

4) Phase-Lock Loop Errors: Phase-lock errors are actual mistakes and can usually be corrected: however, we will mention them anyway. A possible source of error is to lock on a loop-oscillation sideband. The least offset likely in this way would be comparable with the natural frequency of the loop, typically from 10 kHz upwards. Such an error is relatively easily resolved in the more accurate experiments and thus will generally be detected by the experimenter. Another problem arises from spurious modulation of weak signals used to phase-lock oscillators. For example, silicon-controlled rectifer spikes can cause intermittent unlock for say half-a-line-frequency period which could alter the mean locked frequency over a typical 1-s count-averaging time [46]. Careful spectrum analyzer checks are required in such circumstances. The direction of the error is, however, usually random so that accuracy increases with repeated measurement.

V. Summary

Many hundreds of laser frequencies and stabilization reference frequencies have now been measured, and accurate measurements have even been extended as high as the visible, yellow–green, portion of the spectrum.

This has permitted the recent (1983) redefinition of the meter fixing the value of c_0 exactly, so that length and wavelength are based on the same physical standard as time and frequency. Thus length is now referred to the most accurate physical standard available—the cesium clock. Substantial immediate improvements in realizing the meter have been obtained. Also, in such activities as time-of-flight ranging in space, the conversion from time to length does not increase the uncertainty.

Laser frequency measurement spans a spectral range from about 0.5–500 THz. These three orders of magnitude are comparable with the entire frequency span of microwave measurements 0.5–500 GHz; or, in a linear scale, frequency measurements represent a thousand-fold increase. This is extremely significant for communications purposes. The extension of frequency measurements to higher frequencies and improved accuracies is especially important for future experiments to seek frequency standards better than cesium.

The methane clock, current fundamental work towards new frequency standards using heavy molecules in the 10-μm band, and work on trapped ions at visible or higher frequencies [99] all point towards possible frequency standards with reproducibilities and accuracies exceeding that of the current Cs frequency standard.

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REFERENCES

[1] T. H. Maiman, "Stimulated optical radiation in ruby," *Nature*, vol. 187, pp. 493–494, 1960.
[2] A. Javan, W. R. Bennett, Jr., and D. R. Harriott, "Population inversion and continuous optical maser oscillation in a gas discharge containing a He–Ne mixture," *Phys. Rev. Lett.*, vol. 6, pp. 106–110, 1961.
[3] A. D. White and J. D. Rigden, "Continuous gas maser operation in the visible," *Proc. IRE.*, vol. 50, p. 1697, 1962.
[4] A. Crocker, H. A. Gebbie, M. F. Kimmitt, and L. E. S. Mathias, "Stimulated emission in the far infrared," *Nature*, vol. 201, pp. 250–251, 1964.
[5] L. O. Hocker, A. Javan, D. Ranochandra Rao, L. Frenkel, and T. Sullivan, "Absolute frequency measurement and spectroscopy of gas laser transitions in the far infrared," *Appl. Phys. Lett.*, vol. 10, pp. 147–149, 1967.
[6] G. Jones and W. Gordy, "Extension of submillimetre wave spectroscopy below a half millimetre wavelength," *Phys. Rev.*, vol. 135A, p. 295, 1964.
[7] K. D. Froome, "A new determination of the free-space velocity of electromagnetic waves," *Proc. Roy. Soc., Ser. A*, vol. 247, pp. 109–122, 1958.
[8] E. Bergstrand, "A determination of the velocity of light," *Arkiv for Fysik*, vol. 2, 119–150, 1950.
[9] K. M. Evenson, J. S. Wells, L. M. Matarrese, and L. B. Elwell, "Absolute frequency measurements on the 28 and 78 μm cw water vapour laser lines," *Appl. Phys. Lett.*, vol. 16, pp. 159–162, 1970.
[10] D. G. McDonald, A. S. Risley, J. D. Cupp, K. M. Evenson, and J. R. Ashley, "Four-hundredth-order harmonic mixing of microwave and infrared laser radiation using a Josephson junction and a maser," *Appl. Phys. Lett.*, vol. 20, pp. 296–299, 1972.
[11] T. G. Blaney and D. J. E. Knight, "Direct 825th harmonic mixing of a 1 GHz source with an HCN laser in a Josephson junction," *J. Phys. D*, vol. 7, pp. 1882–1886, 1974.
[12] K. M. Evenson, J. S. Wells, F. R. Petersen, B. L. Danielson, and G. W. Day, "Accurate frequencies of molecular transitions used in laser stabilization: the 3.39 μm transition in CH_4 and the 9.33- and 10.18-μm transitions in CO_2," *Appl. Phys. Lett.*, vol. 22, pp. 192–195, 1973.
[13] R. L. Barger and J. L. Hall, "Pressure shift and broadening of methane line at 3.39 μm studied by laser-saturated molecular absorption," *Phys. Rev. Lett.*, vol. 22, pp. 4–8, 1969.
[14] C. Freed and A. Javan, "Standing-wave saturation resonances in the CO_2 10.6 μm transitions observed in a low-pressure room temperature absorber gas," *Appl. Phys. Lett.*, vol. 17, pp. 53–56, and 541, 1970.
[15] G. R. Hanes and C. E. Dahlstrom, "Iodine hyperfine structure observed in saturated absorption at 633 nm," *Appl. Phys. Lett.*, vol. 14, 362, 1969.
[16] Comite Consultatif pour la Definition du Metre, 5th Session, BIPM, Sevres, France, 1973.
[17] T. G. Blaney, C. C. Bradley, G. J. Edwards, D. J. E. Knight, P. T. Woods, and B. W. Jolliffe, "Absolute frequency measurement of the R(12) transition of CO_2 at 9.3 μm," *Nature*, vol. 244, p. 504, Aug. 24, 1973.
[18] T. G. Blaney, G. J. Edwards, B. W. Jolliffe, D. J. E. Knight, and P. T. Woods, "Absolute frequencies of the methane-stabilized He-Ne laser (3.39 μm) and the CO_2, R(32) stabilized laser (10.17 μm)," *J. Phys. D.*, vol. 9, pp. 1323–1330, 1976, and Nature, vol. 254, no. 5501, pp. 584–585, Apr. 17, 1975.
[19] Ch. J. Bordé, M. Ouhayoun, A. Van Lerberghe, C. Salomon, S. Avrillier, C. D. Cantrell, and J. Bordé, "High resolution saturated spectroscopy with CO_2 lasers. Application to the ν_3 bands of SF_6 and OsO_4," in *Laser Spectroscopy IV* (Series in Optical Sciences, vol. 21), H. Walther and K. W. Rothe, Eds. Berlin: Springer-Verlag, 1979, pp. 142–153.
[20] A. Clairon, A. Van Lerberghe, C. Salomon, M. Ouhayoun, and Ch. J. Bordé, "Towards a new absolute frequency reference grid in the 28 THz range," *Opt. Commun.*, vol. 35, no. 3, pp. 368–372, Dec. 1980.
[21] J. Bordé and Ch. Bordé, "Superfine and hyperfine structures in the ν_3 band of $^{32}SF_6$," *Chem. Phys.*, vol. 71, pp. 417–441, 1982.
[22] R. S. McDowell, C. W. Patterson, N. G. Nereson, F. R. Petersen, and J. S. Wells, "CO_2 laser coincidences with ν_3 of SiF_4 near 9.7 μm," *Opt. Lett.*, vol. 6, no. 9, pp. 422–424, Sept. 1981.
[23] D. A. Jennings, F. R. Petersen, and K. M. Evenson, "Frequency measurement of the 260 THz (1.15 μm) He–Ne laser," *Opt. Lett.*, vol. 4, pp. 129–130, 1979.
[24] G. R. Hanes, "Doubly-resonant intracavity generation of second harmonic of 1153 nm radiation with stabilization on h.f.s. components of $^{127}I_2$," *Appl. Opt.*, vol. 18, pp. 3970–3974, 1979.
[25] K. M. Baird, K. M. Evenson, G. R. Hanes, D. A. Jennings, and F. R. Petersen, "Extension of absolute frequency measurements to the visible: frequencies of ten hyperfine components of iodine," *Opt. Lett.*, vol. 4, pp. 263–264, 1979.
[26] D. J. E. Knight, G. J. Edwards, P. R. Pearce, and N. R. Cross, "Measurement of the frequency of the 3.39 μm methane-stabilized laser to $\pm$parts in 10^{11}," *IEEE Trans. Instrum. Meas.*, vol. IM-29, pp. 257–264, Dec. 1980.
[27] A. Clairon, B. Dahmani, and J. Rutman, "Accurate absolute frequency measurements on stabilized CO_2 and He–Ne infrared lasers," *IEEE Trans. Instrum. Meas.*, vol. IM-29, pp. 268–272, 1980.
[28] V. P. Chebotayev, "Optical time scale," *J. Phys.* (*Colloq. C8*), supplement to vol. 42, no. 12, pp. C8-505–C8-512, Dec. 1981.
V. G. Gol'dort, V. F. Zakhar'yash, V. M. Klement'ev, M. V. Nikitin, B. A. Timchenko, and V. P. Chebotaev, *Pis'ma Zh. Eksp. Fiz.*, vol. 8, pp. 157–161, 1982, *transl*: "Development of an optical time scale," *Sov. Tech. Phys. Lett.*, vol. 8, no. 2, pp. 68–69, 1982.
[29] Yu. S. Domnin, N. B. Koshelyaevskii, V. M. Tatarenkov, and P. S. Shumyatskii, *Pis'ma Zh. Eksp. Teor. Fiz.*, vol. 30, no. 5, pp. 273–275, Sept. 5, 1979, *transl*: "Absolute measurements of IR laser frequencies," *JETP Lett.*, vol. 30, no. 5, pp. 253–255, Sept. 5, 1979.
[30] Yu. S. Domnin, N. B. Koshelyaevskii, V. M. Tatarenkov, and P. S. Shumyatskii, *Pis'ma Zh. Eksp. Teor. Fiz.*, vol. 34, no. 4, pp. 175–178, Aug. 20, 1981, *transl*: "Measurement of the frequency of a He–Ne/CH_4 laser," *JETP Lett.*, vol. 34, no. 4 pp. 167–170, Aug. 20, 1981.
[31] C. R. Pollock, D. A. Jennings, F. R. Petersen, J. S. Wells, R. E. Drullinger, E. C. Beaty, and K. M. Evenson, "Direct frequency measurements of transitions at 520 THz (576 nm) in iodine, and 260 THZ (1.15 μm) in neon," *Opt. Lett.*, vol. 8, pp. 133–135, 1983.
[32] 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, "Direct frequency measurement of the I_2-stabilized He–Ne 473 THz (633 nm) laser," *Opt. Lett.*, vol. 8, pp. 136–138, 1983.
[33] V. M. Klementyev, Yu. A. Matyugim, and V. P. Chebotayev, *Pis'ma Zh. Ekp. Teor. Fiz.*, vol. 24, no. 1, pp. 8–12, July 5, 1976, *transl.*: "Mixing of the frequencies 88.37 THz (λ = 3.39 μm), 125.13 THz (λ = 2.39 μm), and 260.1 THz (λ = 1.15 μm) in a gas, and production of continuous coherent emission with combined frequency 473.6 THz (λ = 0.63 μm)," *JETP Lett.*, vol. 24, no. 1, pp. 5–8, July 5, 1976.
V. P. Chebotayev, V. M. Klementyev, Yu. G. Kolpakov, and Y. A. Matyugin, "Frequency synthesis of 0.63 nm radiation by mixing optical frequencies in gas and nonlinear crystals," in *Proc. 2nd Frequency Standards and Metrology Symp.* (Copper Mountain, CO, July 5–7, 1976. Boulder, CO: Nat. Bur. Stand., Frequency and Time Standards Section, Nov. 1976), pp. 368–384.
[34] *Comptes Rendus des Séances de la 17^e CGPM*, BIPM, Sevres,

France, 1983.
[35] BIPM, "Documents concerning the new definition of the metre," *Metrologia*, vol. 19, pp. 163–177, 1984.
[36] S. N. Bagayev, B. D. Borisov, V. G. Gol'dort, A. Yu. Gusev, A. S. Dychkov, V. F. Zakharyash, V. M. Klementyev, M. U. Nikitin, B. A. Tinchenko, V. P. Chebotayev, and V. V. Yumin, USSR Academy of Sciences, Siberian Branch, Institute of Thermophysics, Preprint 78-82, Novosibirsk, 1982, publ. "Optical time standard," *USSR Acad. Sci. Avtometriya*, no. 3, pp. 37–58, May–June 1983, *transl*: Automatic Monitoring and Measurement (9B).
[37] D. J. E. Knight and P. T. Woods, "Application of nonlinear devices to optical frequency measurement," *J. Phys. E.*, vol. 9, no. 11, pp. 898–916, 1976.
[38] V. M. Klement'ev, Yu. A. Matyugin, and V. P. Chebotaev, *Kvant. Elecktron.*, vol. 5, no. 8, pp. 1671–1681, Aug. 1978, *transl*: "Frequency measurements in the optical range: Current status and prospects," *Sov. J. Quantum Electron.*, vol. 8, no. 8, pp. 953–958, Aug. 1978.
[39] L. O. Hocker, D. R. Rao, and A. Javan, "Absolute frequency measurement of the 190 μm and 194 μm gas laser transitions," *Phys. Lett.*, vol. A-24, pp. 690–691, 1967.
[40] H. R. Fetterman, B. J. Clifton, P. E. Trannenwald, and C. D. Parker, "Submillimeter detection and mixing using Schottky diodes," *Appl. Phys. Lett.*, vol. 24, pp. 70–72, 1974.
[41] K. M. Evenson, unpublished.
[42] L. O. Hocker, J. G. Small, and A. Javan, "Extension of absolute frequency measurements to the 84 μm range," *Phys. Lett.*, vol. A-29, pp. 321–322, 1969.
[43] T. G. Blaney, N. R. Cross, D. J. E. Knight, G. J. Edwards, and P. R. Pearce "Frequency measurement at 4.25 THz (70.5 μm) using a Josephson harmonic mixer and phase-lock techniques," *J. Phys. D: Appl. Phys.*, vol. 13, pp. 1365–1370, 1980.
[44] C. O. Weiss and A. Godone, "Harmonic mixing and detection with Schottky diodes up to the 5 THz range," *IEEE, J. Quatum Electron*, vol. QE-20, pp. 97–99, 1984.
[45] K. M. Evenson, D. A. Jennings, F. R. Petersen, and J. S. Wells, "Laser frequency measurements: A review, limitations, extension to 197 THz (1.5 μm)," in *Laser Spectroscopy III*, J. L. Hall and J. L. Carlsten, Eds. Berlin, Germany: Springer-Verlag, 1977, pp. 56–58.
[46] D. G. McDonald, F. R. Petersen, J. D. Cupp, B. L. Danielson, and E. G. Johnson, "Josephson junctions at 45 times the energy-gap frequency," *Appl. Phys. Lett.*, vol. 24, pp. 335–337, 1974.
[47] H-U. Daniel, B. Maurer, and M. Steiner, "A broadband Schottky point contact mixer for visible laser light and microwave harmonics," *Appl. Phys.*, vol. B-30, pp. 189–193, 1983.
[48] R. E. Drullinger, K. M. Evenson, D. A. Jennings, F. R. Petersen, J. C. Bergquist, L. Burkins, and H. U. Daniel, "2.5 THz frequency difference measurements in the visible using metal-insulator-metal diodes," *Appl. Phys. Lett.*, vol. 42, pp. 137–138, 1983.
[49] S. W. Wang, *Lasers and Applications*, vol. III, no. 10, p. 6 (late news), Oct., 1984; see also; S. Y. Wang, D. M. Bloom, and D. M. Collins, "20-GHz bandwidth GaAs photodiode," *Appl. Phys. Lett.*, vol. 42, no. 2, pp. 190–192, Jan. 15, 1983.
[50] A. Chu, H. R. Fetterman, D. D. Peck, and P. E. Tannerwald, "Heterodyne experiments from millimetre wave to optical frequencies using GaAs MESFETs above f_T," in *IEEE Conf. Preprint* (Dallas MTT, 1982), pp. 25–28, 1982.
[51] K. J. Siemsen and H. D. Riccus, "Experiments with point-contact diodes in the 30-130 THz frequency region," *Appl. Phys.*, vol. A35, pp. 177–187, 1984.
[52] H. D. Riccus and K. J. Siemsen, "Point-contact diodes," *Appl. Phys.*, vol. A35, pp. 67–74, 1984.
[53] K. M. Evenson, M. Inguscio, and D. A. Jennings, "Point-contact diode at laser frequencies," *J. Appl. Phys.*, vol. 57, pp. 956–960, 1985.
[54] H. R. Fetterman, P. E. Tannenwald, B. J. Clifton, C. D. Parker and W. D. Fitzgerald, "Far-IR heterodyne radiometric measurements with quasioptical Schotty diode mixers," *Appl. Phys. Lett.*, vol. 33, pp. 151–154, 1978.
[55] F. L. Walls, and A. DeMarchi, "RF spectrum of a signal after frequency multiplication; measurement and comparison with a simple calculation," *IEEE Trans. Instrum. Meas.*, vol. IM-24, pp. 210–217, 1975.
[56] T. G. Blaney, N. R. Cross, and D. J. Knight, "Harmonic mixing and frequency measurements at 2.5 THz using Josephson junctions," *J. Phys. D*, vol. 9, pp. 2175–2180, 1976.
[57] J. S. Wells, "A stabilized HCN laser for infrared frequency synthesis," *IEEE Trans. Instrum. Meas.*, vol. IM-22, pp. 113–118, 1973.
[58] V. G. Gol'dort, V. F. Zakhar'yash, and B. A. Kurnevich, *Pribory i Tekhnika Eksp.*, no. 2, pp. 244–248, Mar.–Apr. 1979, *transl.*: "Wideband unit for phase-frequency locking of lasers," *Instrum. and Exper. Technol.*, no. 2, pp. 557–562, Mar.–Apr. 1979.
[59] S. R. Stein, A. S. Risley, H. Van de Stadt, and F. Strumia, "High speed frequency frequency modulation of far-infrared lasers using the Stark effect," *Appl. Opt.*, vol. 16, pp. 1893–1896, July 1977.
[60] W. R. Leeb, H. K. Phillipp, A. L. Scholtz, and E. Bonek, "Frequency synchronization and phase locking of CO_2 lasers," *Appl. Phys. Lett.*, vol. 41, no. 7, pp. 592–594, Oct. 1, 1982.
[61] D. J. E. Knight, "Optical frequency synthesis at NPL," *J. Phys. (Colloq. C8)*, supplement to vol. 42, no. 12, pp. C8-495–C8-503, Dec. 1981.
[62] J. L. Hall and T. W. Hänsch, "External dye-laser frequency stabilizer," *Opt. Lett.*, vol. 9, no. 11, pp. 502–504, Nov. 1984.
[63] C. Freed, L. C. Bradley, and R. G. O'Donnell, "Absolute frequencies of lasing transitions in seven CO_2 isotopic species," *IEEE J. Quantum Electron.*, vol. QE-16, pp. 1195–1206, 1980.
[64] T. Y. Chang and T. J. Bridges, "Laser action at 452, 496 and 541 μm in optically pumped CH_3F," *Opt. Commun.*, vol. 1, no. 9, pp. 423–426, Apr. 1970.
[65] D. J. E. Knight, "Ordered list of far-infrared laser lines (continuous, lambda > 12 μm)," NPL Rep. QU, 45, 1982, National Physical Laboratory Teddington, Middlesex. TW11 OLW, UK, Feb. 1981, reprinted with added pump-ordered list Sept. 1982. ——, "Far-infrared lasers," in *Gas Lasers* (vol. II of *Handbook Series of Laser Science and Technology*), M. J. Webber, Ed. Boca Raton, FL: CRC Press, 1982, pp. 421–491. The latter list is ordered separately by lasing atom or molecule.
[66] M. Inguscio, G. Moruzzi, K. M. Evenson, and D. A. Jennings, a review paper on frequency measurements of optically pumped FIR laser lines, in preparation, for *Appl. Phys. Rev.*
[67] A. Clairon, B. Dahmani, A. Filimon, and J. Rutman, "Precise frequency measurements of CO_2/OsO_4 and He-Ne/CH_4 stabilized lasers," *IEEE Trans. Instrum. Meas.*, vol. IM-34, pp. 265–268, June 1985.
[68] D. J. E. Knight, G. J. Edwards, P. R. Pearce, K. I. Pharaoh, G. P. Barwood, N. R. Cross, Fan Xi-Sheng, and J. A. Golby, "Progress in an experiment to measure the frequency of an I_2-stabilized dye laser at 520 THz (576 nm)," presented at the VIIIth Vavilov Conf., Novosibirsk, USSR, July 26–28, 1984, submitted to *Izvestia Akad. Nauk SSSR* (Seriya Fizicheskaya).
[69] B. G. Whitford, "Simultaneous phase-lock of five CO_2 lasers to a primary Cs frequency standard," *Appl. Phys.*, vol. B-35, pp. 119–122, 1984.
[70] ——, "Measurement of the absolute frequencies of CO_2 laser transitions by multiplication of CO_2 laser difference frequencies," *IEEE Trans. Instrum. Meas.*, vol. IM-29, no. 3, pp. 168–176, Sept. 1980.
[71] Z. Bay, G. G. Luther, and J. A. White, "Measurement of an optical frequency and the speed of light," *Phys. Rev. Lett.*, vol. 29, pp. 189–192, 1972.
[72] K. M. Baird, H. D. Riccius, and K. J. Siemsen, "CO_2 wavelengths and the velocity of light," *Opt. Commun.*, vol. 6, pp. 91–95, 1972.
[73] J. D. Cupp, B. L. Danielson, G. W. Day, L. B. Elwell, K. M. Evenson, D. G. McDonald, L. O. Mullen, F. R. Petersen, A. S. Risley, and J. S. Wells, "The speed of light: Progress in the measurement of the frequency of the methane stabilized He-Ne laser at 3.39 μm," in *Proc. Conf. on Precision Electromagnetic Measurements* (Boulder, CO, USA), pp. 79–80, 1972.
[74] K. M. Evenson, J. S. Wells, F. R. Petersen, B. L. Danielson, G. W. Day, R. L. Barger, and J. L. Hall, "Speed of light from direct frequency and wavelength measurements of the methane stabilized laser," *Phys. Rev. Lett.*, vol. 29, p. 1346, 1972.
[75] T. G. Blaney, C. C. Bradley, G. J. Edwards, B. W. Jolliffe, D. J. E.

Knight, W. R. C. Rowley, K. C. Shotton, and P. T. Woods, "Measurement of the speed of light," *Nature*, vol. 251, no. 5470, p. 46, Sept. 6, 1974.

[76] T. G. Blaney, C. C. Bradley, G. J. Edwards, B. W. Jolliffe, D. J. E. Knight, W. R. C. Rowley, K. C. Shotton, and P. T. Woods, "Measurement of the speed of light I. Introduction and frequency measurement of a carbon dioxide laser," and "II. Wavelength measurement and conclusion," *Proc. Roy. Soc. Lond. A*, vol. 355, pp. 61–88, and 89–114, 1977.

[77] H. P. Layer, R. D. Deslattes, and W. G. Schweitzer, Jr., "Laser wavelength comparison by high resolution interferometry," *Appl. Opt.*, vol. 15, pp. 734–743, 1976.

[78] J. P. Monchalin, M. J. Kelly, J. E. Thomas, N. A. Kurnit, A. Szoke, A. Javan, F. Zermike, and P. H. Lee, "Determination of the speed of light by absolute wavelength measurement of the R(14) line of the CO_2 9.4 μm band and the known frequency of the line," *Opt. Lett.*, vol. 1, pp. 5–7 and 140, 1977.

[79] J. P. Monchalin, M. J. Kelly, J. E. Thomas, N. A. Kurnit, A. Szoke, F. Zernike, P. H. Lee, and A. Javan, "Accurate laser wavelength measurement with a precision two-beam scanning Michelson interferometer," *Appl. Opt.*, vol. 20, no. 5, pp. 736–757, Mar. 1, 1981.

[80] F. R. Petersen, D. G. McDonald, J. D. Cupp, and B. L. Danielson, "Laser spectroscopy," in *Proc. Vail (CO) Conf.*, R. G. Brewer and A. Mooradian, Eds. New York: Plenum, 1973, pp. 555–569.

[81] P. T. Woods, K. C. Shotton and W. R. C. Rowley, "Frequency determination of visible laser light by interferometric comparison with upconverted CO_2 laser radiation," *Appl. Opt.*, vol. 17, no. 7, pp. 1048–1054, Apr. 1, 1978. *Note*: The speed of light uncertainty excludes a contribution that arises from the practical variation between laboratories in realizing the meter from its definition using the ^{86}Kr lamp. With this contribution, about ± 4 parts in 10^9, the uncertainty becomes ± 1.2 m/s.

[82] K. M. Baird, D. S. Smith, and B. G. Whitford, "Confirmation of the currently accepted value 299 792 458 metres per second for the speed of light," *Opt. Commun.*, vol. 31, pp. 367–368, 1979.

[83] V. M. Tatarenkov, V. G. Il'in, V. I. Kiparenko, V. K. Korobov, and S. B. Pushkin, *Izmeritel'naya Teknika*, no. 2, pp. 15–19, Feb. 1980, *transl.*: "Current state and prospects for frequency measurement in the optical range," *Meas. Techniques (USSR)*, vol. 23, no. 2, pp. 108–114, Feb. 1980.

[84] V. P. Kapralov, G. M. Malyshev, P. A. Pavlov, V. E. Privalov, Ya. A. Fovanov, and I. Sh. Etsin, *Opt. Spektrosk.*, vol. 50, pp. 67–72, Jan. 1981, *transl.*: "Measurement of the wavelength ratio of lasers stabilized by saturated absorption in iodine and methane," *Opt. Spectrosc. (USSR)*, vol. 50, no. 1, pp. 34–37, Jan. 1981.

[85] W. R. C. Rowley and A. J. Wallard, "Wavelength values of the 633 nm laser, stabilized with $^{127}I_2$ saturated absorption," *J. Phys.*, vol. E6, pp. 647–652, 1973.

[86] W. G. Schweitzer, Jr., E. G. Kessler, Jr., R. D. Deslattes, H. P. Layer, and J. R. Whetstone, "Description, performance, and wavelengths of iodine stabilized lasers," *Appl. Opt.*, vol. 12, pp. 2927–2938, 1973.

[87] G. R. Hanes, K. M. Baird, and J. DeRemigis, "Stability, reproducibility, and absolute wavelength of a 633 nm He–Ne laser stabilized to an iodine hyperfine component," *Appl. Opt.*, vol. 12, pp. 1600–1605, 1973.

[88] R. L. Barger and J. L. Hall, "Wavelength of the 3.39 μm laser-saturated absorption line of methane," *Appl. Phys. Lett.*, vol. 22, pp. 196–199, 1973.

[89] K. M. Baird, D. S. Smith, and W. E. Berger, "Wavelengths of the CH_4 line at 3.39 μm," *Opt. Commun.*, vol. 7, pp. 107–109, 1973.

[90] F. R. Petersen, D. G. McDonald, J. D. Cupp, and B. L. Danielson, "Rotational constants for $^{12}C^{16}O_2$ from beats between Lamb-dip-stabilized lasers," *Phys. Rev. Lett.*, vol. 31, pp. 573–576, 1973.

[91] F. R. Petersen, D. G. McDonald, J. D. Cupp, and B. L. Danielson, "Accurate rotational constants, frequencies, and wavelengths from $^{12}C^{16}O_2$ lasers stabilized by saturated absorption," in *Laser Spectroscopy*, R. G. Brewer and A. Mooradian, Eds. New York: Plenum, 1973, pp. 555–569.

[92] J. L. Hall, "Problems with atomic frequency standards of extreme accuracy and a new atomic beam interaction geometry," *Opt. Commun.*, vol. 18, no. 1, pp. 62–63, July 1976.

[93] Yu. S. Domhin, N. B. Koshelyaevskii, V. M. Tatarenkov, and P. S. Shumyatsky, "Precise frequency measurements in submillimetre and infrared region," *IEEE Trans. Instrum. Meas.*, vol. IM-29, pp. 264–267, 1980.

[94] V. P. Chebotayev, "Optical time scale," *J. de Phys. (Colloq. C8)*, supplement to vol. 42, no. 12, pp. C8-505–C8-512, Dec. 1981.

[95] V. F. Zakhar'yash, V. M. Klement'ev, M. V. Nikitin, B. A. Timchenko, and V. P. Chebotaev, *Zh. Tekh. Fiz.*, vol. 53, no. 11, pp. 2241–2244, Nov. 1983, *transl*: "Absolute frequency measurement of the E-line of methane," *Sov. Phys. Tech. Phys.*, vol. 28, no. 11, pp. 1374–1375, Nov. 1983.

[96] V. P. Chebotayev, V. M. Klementyev, M. V. Nikitin, B. A. Timchenko, and V. F. Zakharyash, "Comparison of frequency stabilities of the Rb standard and of the Ne–Ne/CH_4 laser stabilized to the E line in methane," *Appl. Phys. B*, vol. 36, pp. 59–61, 1985.

[97] G. P. Barwood and W. R. C. Rowley, "Characteristics of a $^{127}I_2$-stabilized dye laser at 576 nm," *Metrologia*, vol. 20, pp. 19–23, 1984.

[98] D. J. E. Knight, "A tabulation of absolute laser frequency measurements," Preprint, Aug. 1985 (submitted to *Metrologia*).

[99] D. J. Wineland, W. M. Itano, J. C. Bergquist, J. J. Bollinger, and H. Hemmati, "Frequency standard research using stored ions," *Progr. Quant. Electron.*, vol. 8, pp. 139–142, 1984.