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THE FREQUENCY MEASUREMENT OF VISIBLE LIGHT *

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Abstract. - A discussion of the extension of absolute frequency measurements to the visible will be given along with some new measurements of visible frequency differences using the MIM diode. Future frequency measurements and the redefinition of the meter will also be discussed.

Introduction. - The first absolute frequency measurement of visible radiation was made about two and one-half years ago [1]. Although it was not performed with high accuracy, it demonstrated the feasibility of direct frequency measurements in the visible. Many standards laboratories in the world are now in the process of making highly accurate absolute visible frequency measurements to establish accurate visible frequency references, which might also serve as length standards in a possible redefinition of the meter. The redefinition of the meter may be considered at the next General Assembly of the International Committee on Weights and Measures. The redefinition proposed by the Consultative Committee for the Definition of the Meter [2] states: "The meter is the length equal to the distance traveled in a time interval of 1/299 792 458 of a second by plane electromagnetic waves in vacuum." With this redefinition, the speed of light, c, will be fixed, and the meter will be realized via a frequency measurement of a stabilized laser. It is the advent of the absolute frequency measurement of visible light with the inherently high accuracy of frequency measurement which has made this redefinition possible. Visible radiation is most important for realizing the meter because of alignment ease and higher accuracy length measurement at these shorter wavelengths.

The extension of absolute frequency measurements to the visible requires first, stable oscillators and, secondly, harmonic generating techniques for frequency synthesis purposes (eventually, the Cs frequency standard must be multiplied some 60 000 times). An alternative to harmonic generation would be a dividing technique; however, the technique proposed by Wineland [3] has not yet been proven operational.

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Stabilized oscillators are provided by lasers locked to atomic and molecular resonances. This field has progressed rapidly in the last decade, and some of the stabilized oscillators are challenging the fundamental cesium standard. The various techniques and the progess made are nicely summarized by Hall [4].

Techniques for harmonic generation have not been as efficient above 1 THz as at microwave frequencies where up to 1000 harmonics have been generated. A maximum of 12 harmonics has been achieved with lasers. Consequently, "chains" of oscillators are used to reach the visible. The most useful harmonic generator-mixer operating between 1 and 200 THz discovered so far is the tungsten-nickel point-contact diode. This same diode has been used as a third-order mixer in the visible at frequency differences up to 2.5 THz (to be described later).

In this paper, we will attempt to review progress in the laser frequency measurement field, present some recent results, and make a few comments about future experiments.

<u>The Measurement of the Frequencies of Various Stabilized Lasers</u>. - Various "well-behaved" lasers have been used to probe narrow atomic and molecular resonances, and the narrowest of these have been used for frequency stabilization. Hence, it is the narrow atomic or molecular transition frequency which is being measured. Three sets of these stabilized lasers have emerged and will be described:

1. The CO_2 laser stabilized to a number of molecular absorptions.
2. The 3.39 μm helium-neon laser stabilized to methane.
3. The various sources of visible coherent light stabilized to iodine.

<u>Carbon Dioxide Frequencies</u>. - The carbon dioxide laser is unique since all of the lines (with the exception of the sequence bands) can be stabilized to Doppler-free absorption features in CO_2 itself by the saturated fluorescence technique [5]. The frequencies of lasing transitions in seven CO_2 isotopes, $^{12}C^{16}O_2$, $^{13}C^{16}O_2$, $^{12}C^{18}O_2$, $^{13}C^{18}O_2$, $^{12}C^{16}O^{18}O$, $^{14}C^{16}O_2$, and $^{14}C^{18}O_2$, have now been measured to within an uncertainty of a few parts in 10^9 [6]. The measurements are related to the Cs standard via chains which measured the absolute frequencies of $R_{II}(10)$ and $R_I(30)$ in $^{12}C^{16}O_2$ [7]. Figure 1 shows good agreement among measurements of $R_I(30)$ made by various standards laboratories throughout the world [7-12]. With the improvement in laser stabilization and in the frequency measurement chains, uncertainties have been reduced to less than two parts in 10^{10} [12]. The NRC measurement is different in that it was made by multiplication of CO_2 laser difference frequencies with only CO_2 lasers in the chain, and the preliminary result here shows the feasibility of this technique [11].

Future improvements in the CO_2 laser reference frequencies will require improvement in the laser stabilization in addition to the frequency measurement.

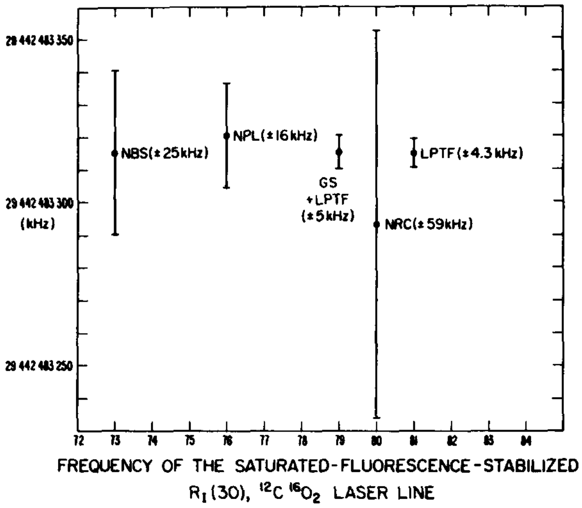

Figure 1. - Frequency of the saturated-fluorescence stabilized $R_I(30)$, $^{12}C^{16}O_2$ laser line. All measurements are uncorrected for baseline slope, pressure, power, or other frequency shifts. The GS + LPTF measurement combines the GS measurement of the OsO_4 stabilized $P_I(14)$ line and the OsO_4 - $P_I(14)$ and $P_I(14)$ - $R_I(30)$ frequency difference measurements by LPTF. The NRC measurement demonstrates use of an all CO_2 laser chain and is a preliminary result.

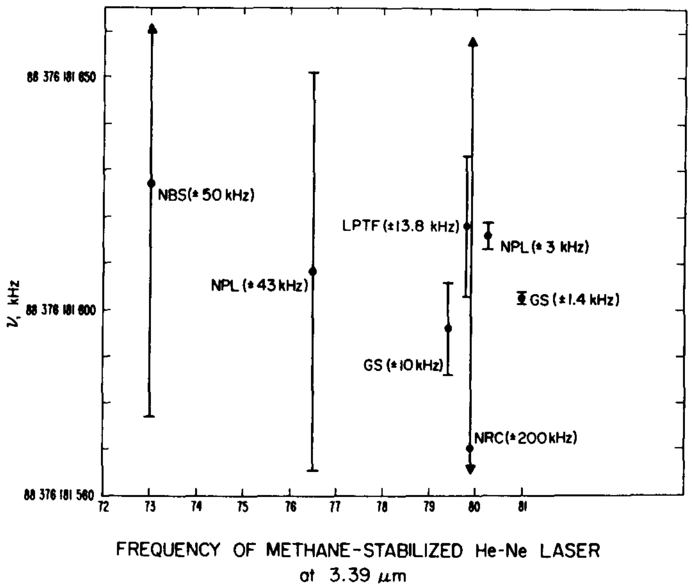

Figure 2. - Frequency measurements of methane-stabilized He-Ne lasers at 3.39 μm. References are: NBS [7], NPL [8], GS [9], LPTF [12], NRC [13], NPL [14], and GS [15], in order.

Selection of heavy molecules such as OsO_4 which have frequency overlaps with CO_2 have provided a thousand-fold reducution in linewidth (1 kHz with a corresponding improveme t in the frequency reproducibility). A judicious selection of absorption lines in these molecules could provide an improved frequency reference grid over the intermediate infrared band [10]. Until that time, the approximately 600 CO_2 laser frequencies known to a few parts in 10^9 are serving well as secondary frequency standards in the infrared.

Methane Frequencies. - The measurements of methane at 88 THz [7-9,12-15] and the uncertainties (1-σ) are shown in figure 2 versus the year each measurement was made. Good agreement between the values is seen, with the discrepancies being due to possible power shifts from the hyperfine structure of methane. The next measurements of this molecule probably will be made on lasers which can resolve the hyperfine structure to eliminate this problem. The smallest reported uncertainty is $\pm 2 \times 10^{-11}$. This frequency measurement is better than best wavelength comparison by about a factor of ten, and hence, this laser could be used as a length standard as is. As was mentioned earlier, however, it is simpler and more accurate to use visible radiation.

Visible Frequencies. - The absolute measurement of the iodine absorption frequency (520 THz) at twice the strong 1.15 μm laser frequency in ^{20}Ne was accomplished with the chain [16] of frequency measurements shown in figure 3. The W-Ni diode was used as the nonlinear element up to 197 THz, but harmonic generation was limited to harmonics of the CO_2 laser in the 88 THz measurement. Subsequently, 148 THz radiation was synthesized in our laboratory in a point-contact diode with five harmonics of a CO_2 laser.

The 260 THz frequency was synthesized by first summing two stabilized CO_2 laser frequencies in the nonlinear crystal, $CdGeAs_2$, and subsequently summing the resultant 5 μm radiation and the 1.5 μm radiation from a ^{20}Ne laser in a $AgAsS_3$ crystal (proustite). The synthesized radiation and the radiation from a Lamb-dip stabilized 1.15 μm ^{20}Ne laser were then mixed on a photodiode and the beat observed on a spectrum analyzer.

The final step [1] was performed in a joint experiment with the National Research Council in Ottawa, Canada in which ten hyperfine transitions in $^{127}I_2$ near 520 THz were measured by comparing the 1.15 μm radiation at one-half the frequency of the I_2 lines with the frequency of the Lamb-dip stabilized pure ^{20}Ne laser at 260 THz. The yellow-green light at 520 THz, generated (in the Hanes NRC laser) by intracavity doubling in $LiNbO_3$ of the 260 THz radiation from a He-Ne laser, was servo-locked to individual hyperfine components of $^{127}I_2$ observed in saturated absorption, and frequencies were determined simply by measurement of the beat frequencies of the two radiations at 260 THz.

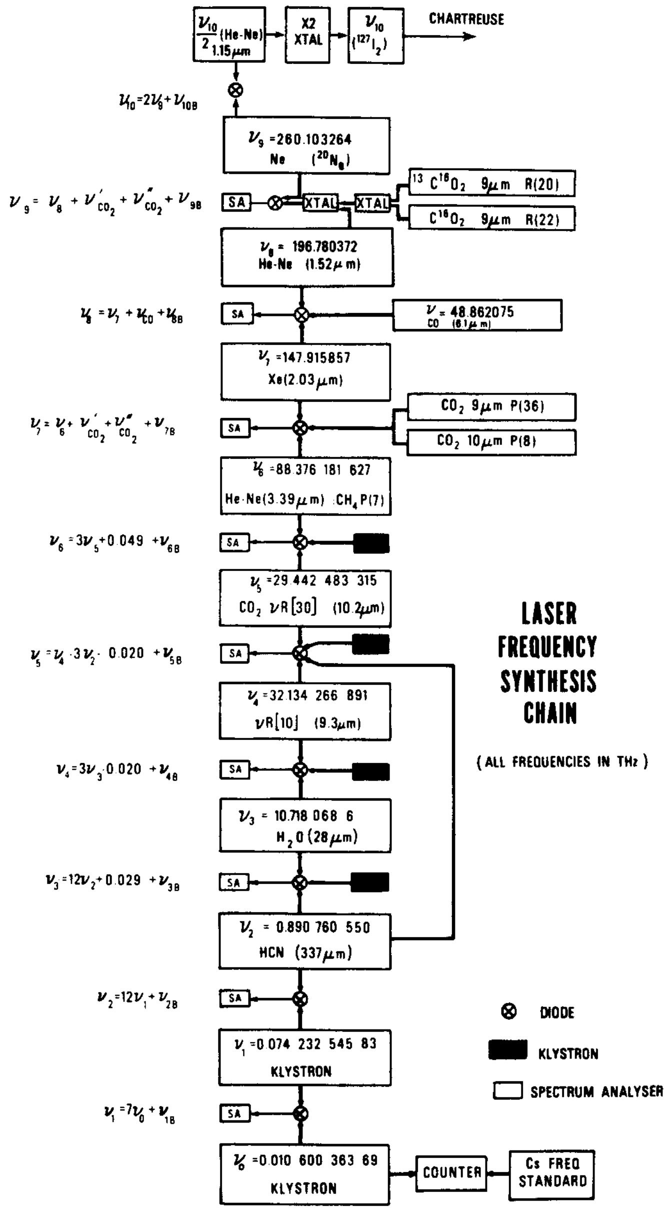

Figure 3. - Old "cesium" to "iodine" frequency chain.

The frequency measurements were done simply by combining the 260 THz beams from the two lasers on a high-speed photodiode and measuring the rf beat between them. The frequency, f_α, of the α-component of the hyperfine structure is given by

$$f_\alpha = 2[f_{Ne} + (f_{beat})].$$

The mean value of the o-component beat frequency was

$$f_{beat} = f_o/2 - f_{Ne} = 154.3 \text{ MHz},$$

and the estimated 1-σ error was 0.5 MHz. The frequency of the o-component of the P(62), 17-1 band of $^{127}I_2$ is thus

$$f_o = 2[f_{Ne} + (f_o/2 - f_{Ne})] = 520\ 206\ 837 \pm 60 \text{ MHz}.$$

A preliminary measurement of the wavelength of this component gave $\lambda = 576\ 294\ 758 \pm 6$ fm [1], from which we calculate $f_o = c/\lambda = 520\ 206\ 811 \pm 6$ MHz. The agreement between the above values for the frequency is satisfactorily within the error limits.

This extension of absolute frequency measurements to the visible paves the way for highly accurate measurements for this portion of the electromagnetic spectrum. The rather large error limit on f_α is due to the free-running 197 THz He-Ne laser used in the measurement of the Lamb-dip stabilized ^{20}Ne, 1.15 μm laser. In this chain, fourteen lasers and six klystrons were used in seven steps, each terminated by a laser actively stabilized to a Doppler-free absorption line when possible.

A simpler chain, shown in figure 4, is now being constructed at NBS, Boulder as an alternative technique for connecting the $^{127}I_2$ stabilized 520 THz laser to the Cs frequency standard. The multiplication factor of 48020 is accomplished with six lasers and four klystrons, a significant reduction from the number in the previous chain. A major change in this chain is the substitution of the color-center laser tuned to half the frequency of the 1.15 μm ^{20}Ne line. This color-center laser, being developed by C. R. Pollock, will use $(F_2^+)_A$ centers in lithium-doped KCℓ pumped with 1.3 μm cw YAG radiation [17]. A ring configuration (shown in figure 5) has been developed, and output powers of 100 mW have been obtained. This chain should have a measurement capability with a fractional frequency uncertainty between 10^{-10} and 10^{-11} for the I_2 transition at 520 THz. A preliminary measurement will be made from the stabilized CO_2 laser to the visible before the final chain is completed. This measurement will be limited by the fractional uncertainty in the CO_2 reference frequency ($\sim \pm 1 \times 10^{-9}$).

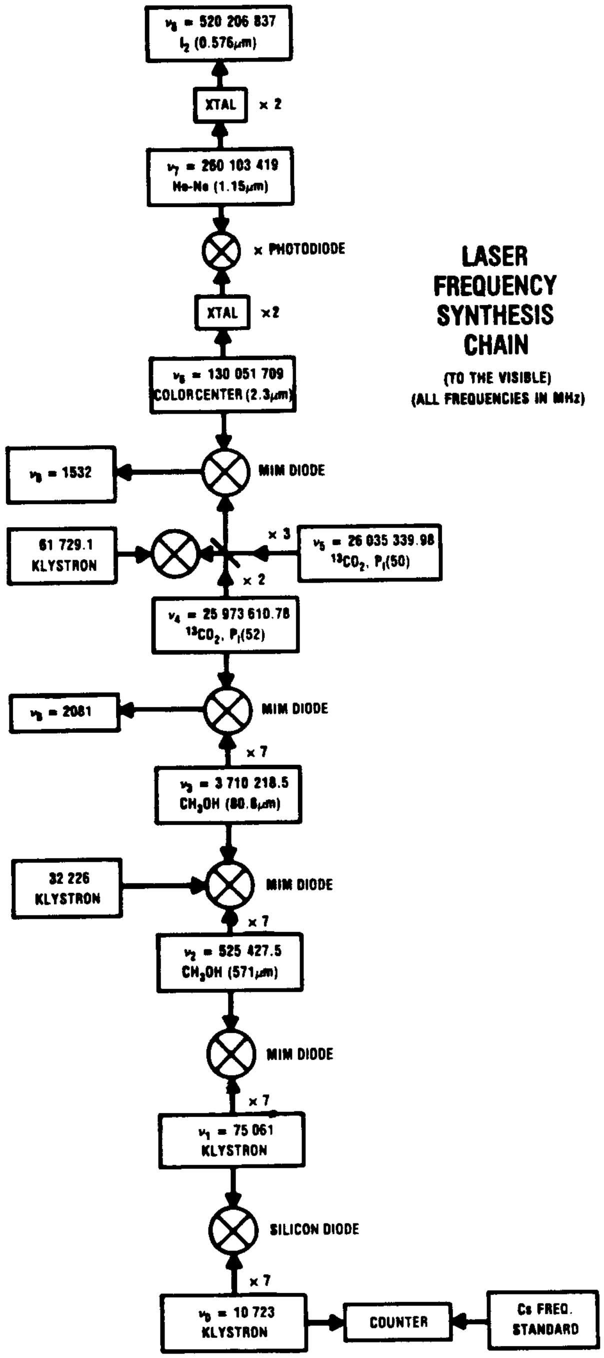

Figure 4. - New "cesium" to "iodine" frequency chain.

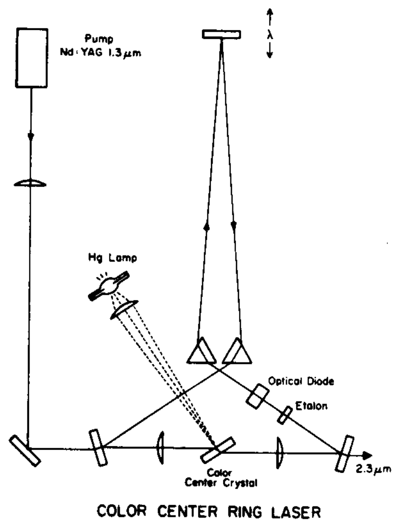

Figure 5 - Color center ring laser.

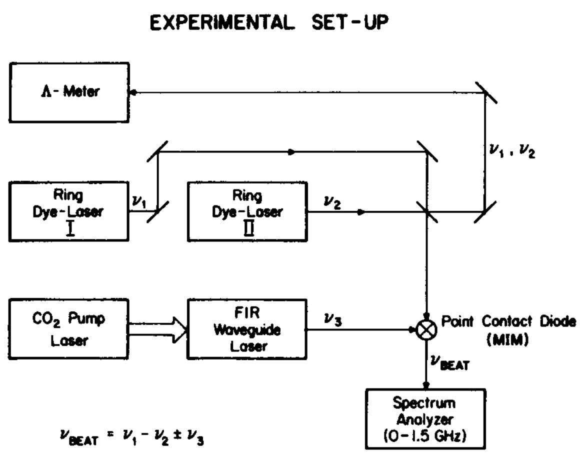

Figure 6.- Experimental arrangement to test the MIM diode in the visible.

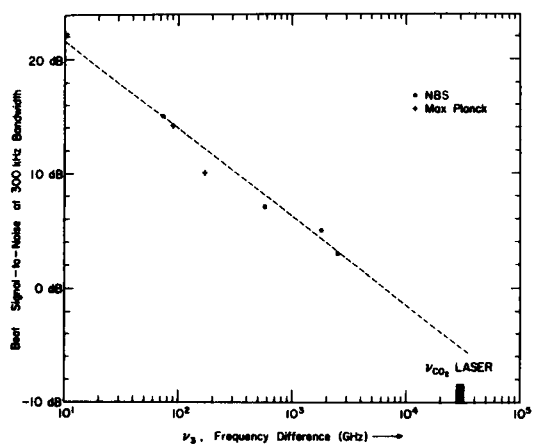

Figure 7. - Visible beat signal-to-noise ratio versus frequency difference from the MIM diode.

This stabilized frequency at 520 THz appears to be an ideal reference from which to synthesize other standard frequencies in the visible spectrum. These frequencies may be synthesized by mixing the 520 THz radiation with an appropriate infrared laser in a nonlinear crystal such as $AgGaS_2$. This crystal has been produced with a wide, low-absorption transmission band ($\alpha < 0.5$ cm^{-1} for $0.5 < \lambda < 10$ μm) and has been used to up-convert 10.6 μm radiation into visible sidebands in the green spectral range. The crystal, however, does not phase match in the entire visible region, and the MIM diode might be used there, as will be described in the next section.

Visible Difference Frequencies Measured at 2.5 THz. - A major breakthrough in the measurement of visible frequency differences occured this year. In a joint experiment between the Max-Planck Institute for Quantum Optics, Garching, W. Germany and the Time and Frequency Division of NBS, Boulder, Colorado, R. E. Drullinger, J. C. Bergquist, D. A. Jennings, F. R. Petersen, Lee Burkins, Ulrich Daniel, and K. M. Evenson demonstrated that the MIM diode (W-Ni or W-Co) could be used to measure the difference frequency between two visible dye lasers separated by as much as 2.5 THz. We also predict the possibility of extending that difference up to 30 THz, i.e., up to the frequency of the CO_2 laser. Before these measurements were made, frequency difference measurements in the visible had been made in three different ways: 1) with photodiodes at difference frequencies up to about 50 GHz, 2) with Schottky barrier diodes at difference frequencies up to 200 GHz, and 3) with nonlinear bulk mixing.

The experiment was inspired by the success of Daniel et al. [18] who used the point-contact diode to measure frequency differences up to 122 GHz. The experimental setup is shown in figure 6. From 0.5 to 2 mW of cw radiation from a far infrared laser was mixed with 15 mW of cw radiation from two different dye lasers which were tuned to a frequency difference approximately equal to the far infrared frequency. Three different far infrared frequencies were used: 0.425, 1.8, and 2.5 THz. Two tunable, stabilized, ring dye lasers were used to generate the two visible frequencies near 575 nm. The spectral linewidth of each was about 200 kHz. Beat signals of a few hundred MHz were observed with a conventional spectrum analyzer. The observed signal-to-noise ratios are plotted in figure 7. Four data points at microwave frequencies are also shown, one at 10 and one at 74 GHz, and two obtained by the Max-Planck group, one at 88 GHz and another at 170 GHz. The signal-to-noise is observed to fall off at 2.3 dB per octave. The reason for this fall-off is not known. To compensate for a possible decrease in coupling efficiency at high frequencies, the power was adjusted to give a constant rectified voltage. Also, the capacitive shunting of the diode is expected to be at least -3 dB per octave, but is not expected to begin falling off until about 7 THz for a 400 Å tip and 30 THz for a 100 Å tip. The radiations from both dye lasers were combined on a beam splitter and focused with a single microscope objective. The wavelength of each dye laser was measured with a λ-meter to about ± 50 MHz. In a final experiment, the dye lasers were tuned 25 THz apart (a CO_2 laser frequency), but a beat note was not observed.

Only the best signal-to-noise ratios were plotted; they varied from 12 to 22 dB at 10 GHz, possibly depending on the sharpness of the diode. Electron micrographs were not taken of the diodes used in the experiments, but previous micrographs have revealed radii varying from less than 100 Å to 600 Å. This variation in tip radius might cause the observed variation in sensitivity.

The experiment was repeated again with much faster stabilization of the dye lasers, which produced a linewidth of less than 10 kHz. However, again, a signal was not observed. In this case, the signal-to-noise obtainable at X-band increased by about 13 dB (a maximum of 35 dB was observed). It was not possible to use one of these "super" diodes tested at 10 GHz for the 25 THz measurement, since during the time required to retune the dye lasers, the contact resistance invariably changed. When the diode was readjusted, it often produced very promising rectified voltages from each radiation, but no beat was observed. With the W-Ni diode, the rectified voltage was generally negative at frequencies below 200 THz, and positive in the visible. The polarity of the rectified voltage from the diode with the cobalt base was less predictable, but this diode performed equally well in the successful difference frequency measurements.

We believe that these experiments were not conclusive and should be repeated under more controlled conditions, i.e., with a measurement of the whisker tip radius and with a check on the absolute accuracy of the λ-meter. We think that the point-contact diode can probably be used in the visible at frequency differences up to 30 THz. This will facilitate the measurement of visible frequency differences in steps of about 30 THz and allow one to obtain a comb of 7 or 8 known frequencies covering the entire visible spectrum.

Conclusion. - Secondary frequency standards extending from the microwave to the visible are becoming available; hence with improvements in the frequency measurement chains, the accuracies of spectral measurements of electromagnetic radiation will soon be limited by the stabilities of the sources themselves. It is also fairly certain that the meter will be defined in terms of the second with the value of c fixed. Thus, the wavelength standard will not be the limitation in the measurement of length as is now the case with the krypton primary standard.

In conclusion, we see that in 20 years, the laser has caused a real revolution in frequency and wavelength metrology as well as in the field of fundamental constants; i.e., one of the fundamental contants (the meter) may soon be defined in terms of another (the second).

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