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Role of Infrared Frequency Synthesis in Metrology

Abstract—Infrared frequency synthesis (IFS) techniques are briefly surveyed, and some important results are summarized. The recent measurement of the frequency of the methane-stabilized He–Ne laser is significant due to the accurate measurement of the methane wavelength and its fundamental role in metrology. The possibilities of an improved value for the speed of light and of additional applications for frequency measurements at various levels of accuracy are discussed.

Frequency synthesis is a technique used by metrologists to generate frequencies and to measure and control frequencies of oscillators. The technique was extended into the infrared in 1967 when the frequency of the HCN laser at 0.891 THz was measured in terms of microwave frequencies [1]. Since that time, infrared frequency synthesis (IFS) has been used to expand frequency measurements upward to a recent value of 88 THz for the methane-stabilized He–Ne laser [2]. It is anticipated that such frequency synthesis will be extended into the visible radiation region within the next few years.

TABLE I

SUMMARY OF SOME INFRARED FREQUENCY SYNTHESIS EXPERIMENTS

Frequency ν_x(THz)	Wavelength (Nominal) λ_x(μm)	Type of Laser	Laser l	 ν_1 (THz)	Laser m	 ν_2 (THz)	Klystron n	 ν_3 (THz)
0.890 760	337	HCN					12	0.0742
0.964 313	311	HCN					13	0.0742
3.821 775	78	H_2O	4	0.964			−1	0.035
3.790 477	79	H_2O	1	3.821			−1	0.031
10.718 073	28	H_2O	12	0.891			1	0.029
28.306 251	10.6	CO_2(P20)	3	10.718	−1	3.821	1	0.027
28.359 800	10.6	CO_2(P18)	3	10.718	−1	3.821	−1	0.026
88.376 245	3.39	He–Ne(CH_4)	3	29.442			1	0.049

To measure an unknown frequency of a laser, one must synthesize a frequency ν_s that is close to the unknown frequency ν_x. The difference between these two frequencies is an intermediate frequency ν_{IF} typically in the 10- to 100-MHz range. The synthesized frequency is

$$\nu_s = \nu_x \pm \nu_{IF} = l\nu_1 + m\nu_2 + n\nu_3$$

where ν_1 and ν_2 are basis laser frequencies determined by prior synthesis experiments, and ν_3 is a microwave frequency. The quantities l, m, and n are harmonic numbers, with m and n allowed both positive and negative values. The harmonic generation, as well as the mixing that produces an intermediate frequency to be measured, occurs in a suitable nonlinear element. Wavelength measurements are sufficiently accurate to determine uniquely the harmonic numbers. The residual uncertainty in measured wavelength requires some searching with a tunable klystron to determine ν_3.

A list of some of the important infrared frequencies and the synthesis schemes employed in some frequency measurements that used CW lasers are shown in Table I.

The HCN laser is the basis laser of lowest frequency in Table I and in Fig. 1. Its frequency was measured by determining the 74-GHz klystron frequency with harmonics of an *X*-band klystron whose frequency in turn was measured with a frequency counter. The HCN laser was then used to measure the 78- and 28-μm water vapor laser frequencies [3], which subsequently formed the bases for measurements at 10 μm [4]. As a result of these measurements at 28 and 10 μm and some previous comparisons at 9 μm [5], the frequencies of approximately 100 lines in various branches of the CO_2 laser were determined [6]. One of these 100 lines forms the basis for the last entry in Table I at 88 THz.

The measurements in Table I were made on free-running lasers and are typically accurate to parts in 10^6, which represents the uncertainty in resetting a laser to the center of its gain curve. At this level of accuracy, the frequency measurements play an important role in infrared spectroscopy. For example, the frequency of the 79-μm line was determined solely because of its role in a laser magnetic resonance experiment [7]. Not only are the previously mentioned laser frequencies available, but bulk nonlinear processes in the near and middle infrared expand the number of well-defined sources enormously. For example, by various combinations of double, triple, and sum mixing of the 100 CO_2 laser lines, one may produce 10^4 to 10^5 signals with well-defined frequencies and with sufficient power for many scientific and measurement applications. The recent demonstrations that microwave sidebands up to 10 GHz [8] can be added to these CO_2 laser lines mean that the large number of discrete frequencies actually become bands of frequencies. Some of these techniques are applicable to lasers in the mid-IR to visible radiation portion of the spectrum.

A second series of measurements designed to synthesize 88 THz with an accuracy of parts in 10^8 or 10^9 is well under way. This is substantially more difficult to accomplish than the first round. To measure accurately the frequency at the 3.39-μm line probably requires that five lasers be stabilized. The lasers are indicated in the block diagram of the synthesis scheme in Fig. 1, along with a possible step for getting to a visible line at 633 nm (6330 Å). At present, some of the lasers have yet to be stabilized for the first time, and none have been stabilized to the degree of the methane-stabilized He–Ne laser [9], [10].

A second difficulty involves the nonlinear element in which the harmonic generation and mixing occur, especially at the upper end of the chain. This nonlinear element is a metal-on-metal diode (see Fig. 2) consisting of a nickel base and a 2.5-μm-diam tungsten cat whisker that also serves as the antenna to

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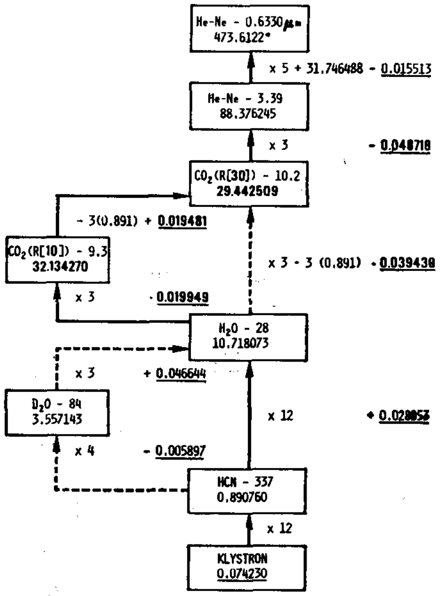

Fig. 1. Laser frequency synthesis chain for measurements leading to a better value for the speed of light. Solid arrows indicate proposed routes, and dashed arrows indicate possible alternatives. Underlined numbers refer to klystron frequencies. The step from 88 THz to 474 THz is a proposed experiment; no frequency synthesis to higher than 88 THz has yet been reported. Wavelengths and frequencies are given in micrometers and terahertz, respectively.

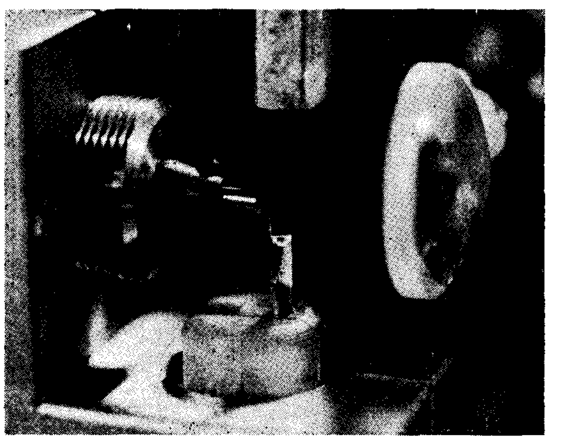

Fig. 2. This diode is the heart of the synthesis scheme in the infrared. The 0.0625-in (1.59-mm)-diam nickel base is held in the tapered section by a set screw. A tungsten cat whisker, so small that it is not visible here, extends upward from the third section of the vertical support, then bends at a right angle and contacts the nickel perpendicularly. On the right is a quartz lens for focusing the 3.39-μm radiation on the diode; above, a waveguide that directs microwave radiation onto the antenna.

bring the terahertz currents to the nonlinear region of the diode. Due to the small dimensions of the antenna, the diode is extremely delicate and short lived. (It is susceptible to air currents and to heat damage due to the laser beam.)

Despite these difficulties, we believe it is now feasible to measure the frequency of the methane-stabilized He–Ne laser with an order of magnitude more accuracy than its wavelength is known. (This becomes especially significant when one considers that the accuracy of the methane wavelength measurement exceeds that now feasible at other frequencies. At longer wavelengths, the accuracy is decreased below the reproducibility of the krypton length standard, i.e., a part in 10^8. This is due to diffraction corrections and to difficulties in fabricating optical components which give accurate results at both the visible and the longer wavelength IR regions of the spectrum. Also, at shorter wavelengths, stabilized lasers with a stability commensurate with the methane-stabilized He–Ne laser do not presently exist.) An accurate frequency measurement of this methane device, combined with the wavelength measurement already performed by Barger [11] and by Giacomo [11], would give a value for the speed of light which would be a factor of 30 more accurate than the present value. A more definitive value for the speed of light is of considerable interest to metrologists and to the scientific community in general.

The third level of accuracy in the frequency measurement work will be to measure the frequency of the methane 3.39-μm line to a part in 10^{10} or 10^{11}. This requires further refinement in the degree of stability of the lasers involved. Recent work of Petersen and Danielson at NBS and Freed and Javan at M.I.T. indicates that the requisite stability is attainable for the CO_2 lasers [12], [13]. The HCN laser has been phase-locked to a microwave chain at NBS and elsewhere [8]. At present no really desirable technique exists for stabilizing the H_2O laser. One would like some molecular absorption stabilization scheme that would permit one to measure the frequency of the 28-μm line accurately, and then use it as a secondary reference. This difficulty at the part in 10^8 level may be circumvented by locking the water vapor laser to the stabilized laser above it in the synthesis scheme in Fig. 1.

Recent developments in the harmonic generator–mixer area could expedite progress at this third and higher levels of accuracy and precision. McDonald and co-workers have recently synthesized to the 3.82-THz frequency of an H_2O laser by using a Josephson junction to generate the 401st harmonic of an X-band signal [14], following earlier success in using the 100th harmonic to synthesize to the 0.89-THz frequency of an HCN laser [15]. New empirical information is being obtained relative to the design and performance of the whisker diodes. Thin-film diodes are being developed by Mullen at NBS as possible substitutes for the point contact diodes.

Although infrared spectroscopy of molecules can make good use of the present accuracy of IFS, there are some interesting applications that will require improved accuracy. As discussed in the following letter [16], the ability to measure the frequency of methane to an accuracy of a part in 10^{11} or better would lead to an additional, fundamental role for IFS. There would then be the opportunity to define the value of the speed of light in conjunction with the adoption of a unified standard for frequency, time, and length.

After additional development, the long-term stability of the relatively simple and inexpensive methane-stabilized He–Ne lasers may possibly become competitive with the stability of cesium-beam atomic clocks (parts in 10^{13} to 10^{14} range). Then, if IFS of comparable accuracy were possible, it would allow these methane devices to be used as stable, calibrated, working (secondary) frequency standards and clocks. Although this development is hypothetical, and might not occur for many years, its impact on timekeeping, frequency metrology, and time-frequency dissemination systems could be considerable.

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