LASER FREQUENCY MEASUREMENTS: A REVIEW, LIMITATIONS, EXTENSION TO 197 THz (1.5μm)

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Abstract

The CO_2 and He-Ne lasers stabilized respectively with CO_2 and CH_4 are now accepted as frequency and wavelength standards at 9 to 12, and 3.39 μm. This is due to their excellent frequency stability and accuracy, and the direct measurement of their frequencies. The measurement of both the frequency and wavelength of the CH_4 device yielded a hundred fold increase in the accuracy of the speed of light. This paper reports the extension of direct frequency measurements to the 197 THz (1.5 μm) cw line of a He-Ne laser, and reviews the current status of laser stabilization and speed of light measurements.

Introduction

With the advent of the laser, coherent sources of radiation became available from the microwave to the visible portion of the electromagnetic spectrum. Thus, one could think of measuring photon energy via either frequency or wavelength metrology techniques. The coherence of the cw laser's radiation is most accurately measured via direct frequency measurements, which do not suffer from the limitations of wavelength measurements: diffraction, mirror curvature, and phase shift at the mirror surface. These limit the accuracy in wavelength measurements to about $\pm 1 \times 10^{10}$ [1]. At present, the absolute comparisons of wavelength are limited by uncertainties in the realization of the definition of the meter to $\pm 4 \times 10^{-9}$ [2]. In contrast, the absolute accuracy of laser frequency measurements is limited presently by the stabilities of lasers used in frequency synthesis. Eventually, the limitation will be in the accuracy of the sources which are now $\pm 0.8 \times 10^{-13}$ for the time standard [3] and $\pm 2 \times 10^{-13}$ for the 3.39 μm CH_4 stabilized He-Ne laser [4]. Most laser frequency measurements, so far, have been made through the use of a non-linear device: a tungsten nickel catwhisker point contact diode. This paper will discuss the stability of laser sources; the techniques of laser frequency measurement; the non-linear devices used in laser frequency measurements; laser speed of light measurements; the redefinition of the meter; the extension of laser frequency measurement to 197 THz; and some limitations of the techniques used so far in laser frequency measurements.

Stabilization of Lasers

Frequency measurements are mankind's most accurate measurements, with the accuracy limited only by the coherence of the sources themselves. For this reason it is appropriate to begin this discussion by considering some of the various types of very coherent sources, specifically those which can be considered as possible frequency standards themselves.

Although the spectral purity of a free running laser has been shown to be as good as a few Hertz [5], without some means of controlling the length of the cavity, the frequency can vary over the Doppler width of the laser transition from a few megahertz in the far infrared to over a thousand megahertz in the visible. (We are specifically discussing Doppler width

instead of gain band width since most stable lasers use a gaseous gain medium.) This same Doppler width generally allows the oscillation of several longitudinal modes in the cavity, so that several frequencies spaced by c/2L, where c is the speed of light and L is the length of the cavity, will be oscillating at one time. To force single frequency oscillation, one generally chooses a laser cavity sufficiently short so c/2L is greater than the gain width. Then the laser frequency can be locked to some reference by controlling the length of the cavity. This reference can be either a frequency synthesized from other stable and known sources, or, some Doppler-free spectral feature. In the latter category, the lasers locked to these "Lamb dip" type of features become independent frequency or wavelength references. The best known of these are: a saturated fluorescence in CO_2 for locking each CO_2 line [6]; a saturated absorption in CH_4 to lock the 3.39 μm (88 THz) line of the He-Ne [7], saturated absorbtion in iodine to lock the 632.8 nm He-Ne [8], and a molecular beam technique to lock the argon laser [9]. The short term line width, the stability, and the accuracy capability of these radiation sources are shown in Table 1.

The two HeNe lasers locked to their respective sources have been compared with the present standard of length sufficiently accurately so that values of their wavelengths have been recommended as secondary standards by the Comite Consultatif pour la Definition du Metre [20]. The values are: methane, P(7), band ν_3; 3. 392 231 40 x 10^{-6} m and, iodine 127, R(127), band 115, component i; 0.632 991 399 x 10^6 m.

For absolute frequency measurements the present standard, Cs, as can be seen in Table 1, is limited to $\pm 8 \times 10^{-14}$. Other sources, such as the hydrogen maser may be more stable, but problems with the wall shift have excluded it from consideration as a primary standard.

The 3.39 μm He-Ne laser, although still in its early stages of development, is second only to Cs in accuracy capability and even surpasses it in stability at 100 sec. Thus, this He-Ne laser is an excellent secondary frequency and wavelength standard in the infrared. However, before a frequency standard at this high frequency can be considered, more efficient and simpler techniques of frequency synthesis must become more commonplace. A great deal of work is underway on new techniques, but what simplification will occur is not yet certain.

Frequency Measurements

Frequencies may be directly counted up to about 500 MHz by timed gating of electronic counters. At higher frequencies, a heterodyne technique is generally used whereby the difference between two nearly equal frequencies is generated and directly counted. One of the frequencies is synthesized from known frequencies by some sort of non-linear device which can either sum two or more different frequencies or generate harmonics, or both. The synthesis at laser frequencies presents special challenges since the frequencies are so high. An excellent review paper summarizes the application of non-linear devices to optical frequency measurement [21]. The silicon diode which generates many different orders of harmonics so well in the microwave ceases to respond at about one terahertz, approximately the frequency of the HCN laser, one of the lowest frequency lasers. Bulk mixers have been used in laser frequency synthesis for some time; however, due to low efficiency in these devices, phase matching must be used; thus, for cw radiation they are limited to 2nd harmonic generation or two waves mixing.

Table 1 Frequency Sources

Source	Ref	λ, μm	ν, THz	Stabilizing Source	Short-Term Line Width Unlocked 10^{-3} sec	Stability 100 sec	Accuracy Capability
H maser	10, 11, 12		0.0014	H. Active		10^{-15}	1×10^{-12}
H maser	13			H. passive		10^{-13}	1×10^{-12}
Cs beam	3		0.0092	Cs beam		7×10^{-14} (estimated)	8×10^{-14}
N_2O laser	14	10–11	27–30	N_2O Sat. Fl.			$\approx1\times10^{-9}$
$^{12}C^{16}O_2$	15	9.1 to 11	27–33	CO_2 Sat. Fl.	2.5×10^{-13}	6×10^{-13}	$\approx10^{-10}$
CO_2 isotope laser	15	9.0 to 12.4	24–33 all isotopes	CO_2 Sat. Fl.	2.5×10^{-13}	6×10^{-13}	$\approx10^{-10}$
He-Ne laser compact intracavity device	16	3.39	88	CH_4 Sat. Ab.			2×10^{-11}
He-Ne	17, 4, 18	3.39		CH_4	2×10^{-13}	5×10^{-15}	2×10^{-13}
He-Ne laser	19	0.63	475	I_2 Sat. Ab.		10^{-12}	$\approx10^{-10}$
Ar laser	7	0.5145	550	I_2 beam	4×10^{-8}	2×10^{-13}	1×10^{-11}

Another device, a MIM (metal-insulator-metal) diode, first used at laser frequencies in A. Javan's laboratory [22], similar in use to the silicon diodes used in the microwave region, is the main device used in laser frequency synthesis. It is similar to the catwhisker diode used in the early radio receivers; however, it uses only metals and the natural oxide that forms in the nickel base as the insulator. It has been used to generate as high a harmonic as the twelfth with an HCN laser [23] and has been used to generate harmonics to 148 THz. Later in this paper, we will describe its use as a 3rd and 4th order mixer at 197 THz (1.52 μm).

The catwhisker is fabricated from a 10 to 25 μm diameter tungsten wire electrochemically sharpened to a tip radius of about 0.05 μm. The straight portion of the whisker from the tip to a right angle bend serves as the antenna [24] to concentrate the focused laser field at the tip where the non-linear processes occur at the 1nm natural nickel oxide layer.

The unknown frequency, ν_x, is determined from the synthesis obtained when all of the lower frequency radiations impinge on the diode. That is: $\nu_x = l\nu_1 \pm m\nu_m \pm n\nu_n \pm \nu_b$ where ν_1, ν_m, and ν_n are known frequencies, 1, m, and n are the harmonics, and ν_b is the RF beat frequency. The sum, 1+m+n+1 is the mixing order. The signal to noise ratios obtainable with this device are shown in Fig. 1. These are obtained with single frequency
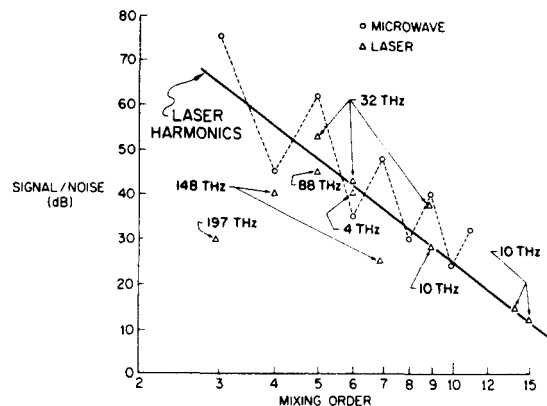

Fig. 1. Signal to noise ratios obtained with tungsten-nickel MIM diodes as a function of mixing order and at various frequencies between 10 and 1000 MHz with a bandwidth of 30 or 100 KHz, a scan width of 200 or 500 KHz/cm, a scan time of 2 ms/cm, and a video filter of 1 KHz.

lasers with outputs of about 100 mw. They were taken with a standard spectrum analyser at between 10 and 1000 MHz with a bandwidth of 30 to 100 kHz, a scan width of 200 to 500 kHz/cm, scan time of 2m sec/cm and a video filter of 1 kHz. Some of the details of microwave and lower frequency laser operation are presented in an earlier paper [25]. The signals at higher frequencies were taken with the larger settings on the spectrum analyser; this is necessary at the higher frequencies since a free-running laser's jitter is approximately proportional to its frequency. This accounts for only a part of the fall off at higher frequency.

The most widely accepted explanation of the diode's operation is that the mechanism is due to tunnelling [26, 27], but, there is some evidence for more than one phenomena occurring in the diodes. The impedance of the diode may be adjusted from about 10 to 10,000 Ω by the microadjustment of the contact pressure. The more sensitive contacts are at higher impedances, and the more stable are at the lower impedances. Usually a compromise at about 200 to 600 ohms is made and the diode is then often stable for several days at a time, and with readjustment of the contact, the whisker can be used for several weeks.

Normally (about 95% of the time), and at frequencies less than 100 THz the laser radiation is rectified in the diode and it drives the tungsten whisker negative. Under some conditions, a bias improves the heterodyne signal; however, it usually is not a significant improvement, and consequently, the diode does not have a dc bias applied.

At frequencies above 100 THz, the rectified polarity usually reverses (ie. the tungsten is driven positive). However, by increasing the dc impedance of the diode a negative driven whisker can sometimes be achieved. For example, at 148 THz, an impedance of about 600 ohms is generally necessary for negative rectification. The negative signal is not necessary for harmonic generation, but somewhat higher sensitivity generally accompanies this signal. Although we have observed antenna patterns at wavelengths as short as 2 μm, generally the antenna becomes less effective due to an increasing resistivity of tungsten at these frequencies. Consequently, we have achieved significantly greater signals with the use of high quality, long working distance microscope objectives to focus the radiation at wavelengths of 2 μm and less. Sharp angular dependence is absent, and the radiation is usually focused at 45° with respect to the antenna with the polarization of the laser radiation lying in the plane determined by the whisker and the laser beam.

The generation of third order or higher harmonic signals is the only satisfactory criterion we have found to test the diodes. For example, at 197 THz, the rectified voltage is generally positive, and the positive signal is not critical with respect to the laser polarization, nor is it extremely critical with respect to the focus. However, to obtain a third order signal, an extremely critical focus must be found which is indicated only by a somewhat sharper square wave signal from a 1 kHz chopped laser beam. The heterodyne signal is over an order magnitude larger with the polarization in the plane of the antenna and laser beam. Thin film diodes have been fabricated [26, 28], however, as yet, they have not been tested for high frequency operation (i.e. with third order mixing).

Chebotayev and co-workers [29] used a very clever scheme to achieve 3 wave frequency synthesis of visible radiation which relies upon the non-linear properties of neon itself in the Helium-Neon laser. This unique mixing occurs because of some common energy levels in 4 lasing transitions in Ne: the He-Ne laser can oscillate on the 0.6330 μm, 3.39 μm, the 2.39 μm, and the 1.15 μm lines. The sum of all of the IR energies is exactly the energy of the 0.6330 μm line since the top-most energy level of the 3 IR frequencies and the bottom most are common with the 0.6330 μm energy levels. Therefore, when a prism and separate mirrors for each of the 3 IR lines are installed on the laser, the induced polarization from the sum of the IR radiations induces a visible transition, and about 1 μwatt of 6330 radiation emerges (with no 0.6330 μm mirror on the cavity). The 0.6330 μm radiation

is thus the sum of all 3 IR frequencies. Of the 3 IR frequencies, only that of the 1.15 μm line has not been reached; thus, once the 1.15 μm frequency is measured, synthesis to the visible will have been accomplished.

Alternatively, it is quite easy to double the frequency of the 1.15 μm laser in a lithium niobate crystal; in fact, we have obtained 100 μw of visible light from 30 mw of 1.15 μm radiation.

The Speed of Light

The advent of lasers and in particular the extension of direct frequency measurement into the infrared region, has been responsible for the 100 fold improvement in the accuracy of the measurement of the speed of light. It has taken c from one of the less accurately known fundamental constants and made it the most accurately known. In fact, there is now the definite possibility of fixing the value of c with a redefinition of the meter [2, 30].

We will summarize all recent measurements of c = λυ where c is obtained by the measurement of both the wavelength, λ, and the frequency, υ, of a stabilized laser. Table 2 gives the results of various frequency and wavelength measurements yielding values of c; these values are graphed in Fig. 2. Also listed is the value of c recommended by the CCDM [2]: for use
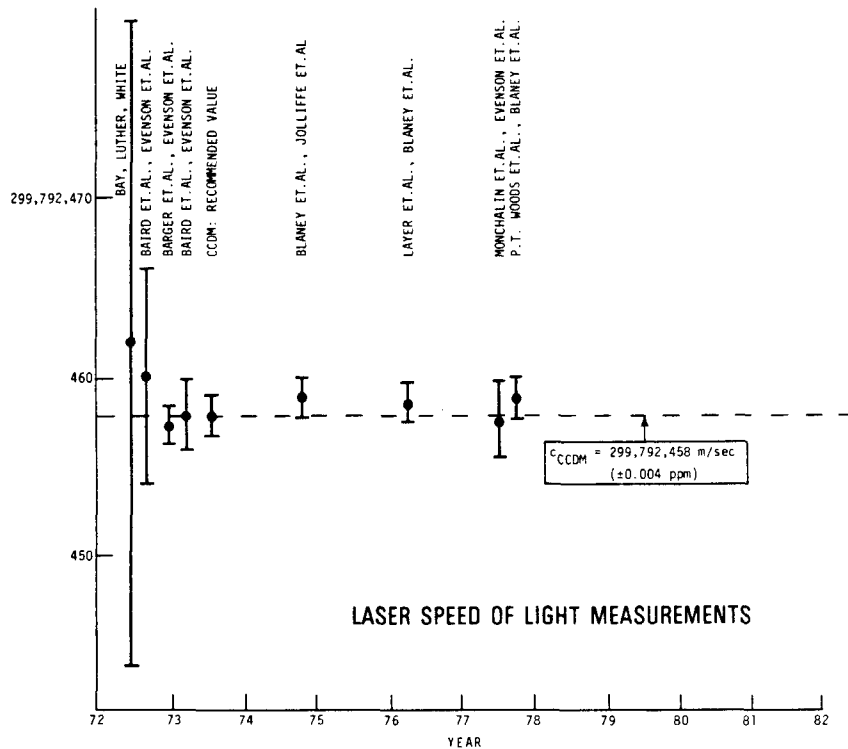
Fig. 2. Values of the speed of light obtained using lasers since 1972.

Table 2. Laser Measurements of c = λν

Laser	Stab.	λ vac., μm[1]	Ref	ν, THz	Ref	c, m/sec
He-Ne	Ne	0.63299147(1)	30	473.612166(29)THz[2]	30	299,792,462(18)
CO_2	CO_2	nine values (9 to 10 μm)	31	nine frequencies	32, 33	299,792,460(6)
He-Ne	CH_4	3.392231390(14)	34, 35	88.376181627D(50)[3]	34, 36	299,792,457.4(1.2)
He-Ne	CH_4	3.392231400(20)	37	88.376181627D(50)[3]	34, 36	299,792,458(2)
CO_2 R(12)	CO_2	9.317246348(44)[4]	38	32.176079482(27)	39	299,792,459.0(1.2)[3]
He-Ne	CH_4	3.392231405(14)[4]	40,1	88.376181608(43)[3]	41	299,792,458.7(1.2)[3]
CO_2 R(14)	CO_2	9.305385613(70)	42	32.217091275(24)	36,43	299,792,457.6(2.2)
CO_2R(12)	CO_2	9.317246340(38)[4,5]	45	32.176079482(27)	39	299,792,458.8(1.2)[3]
CCDM Value 2, 20						299,792,458(1.2)

1. Average of the center of gravity and peak of the asymetrical line of Krypton length standard.
2. In this frequency determination, the optical frequency was not directly counted.
3. A third determination of the frequency of methane with respect to CO_2 is in excellent agreement with the values here and helps confirm these frequencies[44].
4. The uncertainty in the krypton standard ($+4\times10^{-9}$) was added in quadrature to the published uncertainty.
5. This number represents a remeasurement of the wavelength shown in line 5 by the same workers.

in distance measurements where time-of-flight is converted to length; for the fundamental constant; and for use in converting frequency to wavelength. The uncertainty in this value ($\pm 4 \times 10^9$) is the estimated uncertainty in the realization of the definition of the meter itself and results from asymmetries in the line, the line width, and variations from lamp to lamp. This uncertainty has been added to some of the recent determinations of λ (see footnote 3 in Table 2).

All of the values of c except that in row 6 have appeared in the literature; this highly accurate value is independent of the other methane value since both the wavelength and frequency were independently measured. One sees a remarkable convergence of the values of c for the first time in history.

Redefinition of the Meter
As a result of the recommendations made by CCDM, two different definitions of a new length standard must be considered. First, we can continue as before with separate standards for the second and meter, but with the meter defined as the length equal to $1/\lambda$ wavelengths in vacuum of the radiation from a stabilized laser instead of from a ^{86}Kr lamp. Either the methane-stabilized He-Ne laser at 3.39 μm (88 THz) or the I_2-stabilized He-Ne laser at 0.633 μm (474 THz) appears to be suitable candidates. The 3.39 μm laser is already a secondary frequency standard in the infrared, and hopefully, direct measurements of the frequency of the 0.633 μm radiation will give the latter laser the same status in the visible. The 3.39 μm laser frequency is presently known to within 6 parts in 10^{10}, and the reproducibility and long term stability have been demonstrated to be better by more than two orders of magnitude. Hence, frequency measurements with improved apparatus in the next year or two are expected to reduce this uncertainty to a few parts in 10^{11}. A new value of the speed of light with improved accuracy would thus be achievable if the standard of length were redefined in terms of the wavelength of this laser.

Alternately, one can consider defining the meter as a specified fraction of the distance light travels in one second in vacuum (that is, one can fix the value of the speed of light). The meter would thus be defined in terms of the second; and, hence, a single unified standard would be used for frequency, time, and length. What at first sounds like a rather radical and new approach to defining the meter is actually nearly one hundred years old. It was first proposed by Kelvin in 1879 [46], and has been recently reemphasized by Townes [47] and Bay [48]. With this definition, the wavelength of all stabilized lasers would be known to the same accuracy with which their frequencies can be measured. Stabilized lasers would thus provide secondary standards of both frequency and length for laboratory measurements, with the accuracy being limited only by the reproducibility, measurability, and long term stability. It should be noted that, prior to the CCDM value, an adopted nominal value for the speed of light was already in use for high-accuracy astronomical measurements [49]; now however, with the adoption of the CCDM value [2] by the IAU, there is only one standard of length in existence. A definition which fixes c for all users would certainly be desirable from a philosophical point of view.

Extension of Direct Frequency Measurements to 197 THz
The frequency of a cw, free running, 8 m long, 15 to 20 mw, HeNe 1.5 μm laser line was directly measured using both third and fourth order mixing in a tungsten nickel point contact diode. The third order frequency synthesis
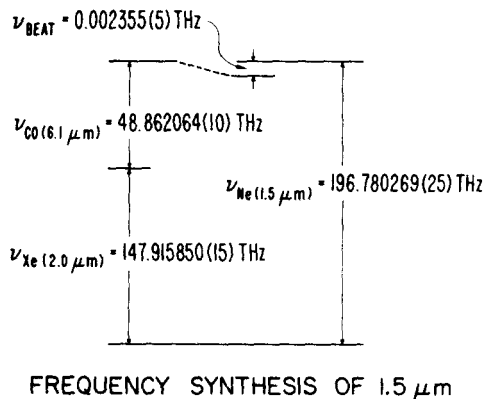
$$\nu_{BEAT} = 0.002355(5)\,THz$$

$$\nu_{CO(6.1\,\mu m)} = 48.862064(10)\,THz$$

$$\nu_{Ne(1.5\,\mu m)} = 196.780269(25)\,THz$$

$$\nu_{Xe(2.0\,\mu m)} = 147.915850(15)\,THz$$

FREQUENCY SYNTHESIS OF 1.5 μm

Fig. 3. Third order synthesis of 197 THz in MIM diode.

scheme is shown in Fig. 3. Forty mw of 148 THz radiation from the Xe laser was focused at 45° with respect to the tungsten antenna on one side of the diode, and the 197 THz was focused at 45° on the other side. Radiation from each of these lasers was isolated from other lines by means of a prism external to the laser. About 100 mw of 49 THz CO laser radiation from the J=14, 19-18 band was focused at about 5° with respect to the antenna. High quality (i.e. expensive) long-working distance microscope objectives were used for the 148 and 197 THz radiations. A 10 db signal to noise beat note at 2.355 GHz was obtained, however a 20 db transmission loss was measured at 2.4 GHz; thus, an estimated signal to noise of 30 db is plotted in figure one. To optimize the signal to noise, the spectrum analyser was set at a band width of 100 kHz, the video filter, at 1 kHz, and the sweep rate, at 5ms/cm.

The frequency obtained by third order mixing was:

$$\nu_{Ne} = 196.780\ 269(25)\ THz = \nu_{2\mu m} + \nu_{CO} + \nu_{beat}$$

where $\nu_{2\mu m}$ = 147.915 850(15) THz[51], ν_{CO} = 48.862 064(10) THz [52], and ν_{beat} = 0.002355(5) THz. Additional uncertainties resulted from setting the Xe and CO lasers to the tops of their free running gain curves, 10 and 5 MHz respectively. The uncertainties were added in quadrature, giving an estimated uncertainty of 25 MHz.

The 4th order experiment was done with a different CO laser line, the J=20 line from the 18-17 band, ν_{CO} = 48.917 212(10) THz; plus the ν_{k1} = 0.051745 THz radiation from a microwave klystron. The result was:

$$\nu_{Ne} = 196.780\ 274(25)\ THz = \nu_{2\ \mu m} + \nu_{CO} + \nu_{k1} + \nu_{beat}.$$

In this case ν_{beat} = 0.001043(5) THz and the signal to noise was 10 db. The frequencies of the CO lines were calculated from the molecular constants of Todd et al [53] and the two resultant Ne frequencies were in much closer agreement than those using earlier CO molecular constants. Each experiment

was repeated 3 times and the results were always within 5 MHz of these average values.

The average of each of these two experiments, thus, gives a value of

$$\nu_{Ne} = 196.780\ 271(25)\ THz,$$

and using the recommended value of c

$$\lambda_{vac} = 1.523\ 4884(2) \times 10^{-6}m.$$

This number is in agreement with that given in spectral tables [53].

The entire frequency chain connecting this laser with the Cs frequency standard is shown in Fig. 4. The 197 THz radiation should be useful in reaching 260 THz (1.15 μm) via

$$\nu_{1.15} = \nu_{1.5} + \nu'_{CO_2} + \nu''_{CO_2} + \nu_{beat}.$$

However; this experiment has been tried unsuccessfully on two different occasions. Diode-burnout with 1.15 μm radiation seemed to be the main problem. It is not too surprising because the reflectivity of tungsten drops to about 60% at 1 μm. It is hoped that gold coating might decrease the burnout problem and allow the continuation of frequency measurements to the visible. Even if this is not the case, we are now close enought to the visible that 2nd harmonic generation or 2 wave mixing in bulk materials will certainly produce direct frequency measurement in the visible soon.

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$\nu_8 = 196.780271$
He-Ne (1.52 μm)

$\nu_8 = \nu_7 + \nu_{CO} + \nu_{8B}$

SA — DIODE — $\nu_{CO} = 48.862064$ (6.1 μm)

$\nu_7 = 147.915850$
Xe(2.03 μm)

$\nu_7 = \nu_6 + \nu_{6,7}' + \nu_{6,7}'' + \nu_{7B}$

SA — DIODE — $\nu_{6,7}'$ CO$_2$ 9 μm P(36)

$\nu_{6,7}''$ CO$_2$ 10 μm P(8)

$\nu_6 = 88.376.181.627$
He Ne(3.39 μm) CH$_4$ P(7)

$\nu_6 = 3\nu_5 + 0.049 + \nu_{6B}$

SA — DIODE — KLYSTRON

$\nu_5 = 29.442.483.315$
CO$_2$ νR[30] (10.2 μm)

$\nu_5 = \nu_4 - 3\nu_2 - 0.020 + \nu_{5B}$

SA — DIODE — KLYSTRON

$\nu_4 = 32.134.266.891$
νR[10] (9.3 μm)

$\nu_4 = 3\nu_3 - 0.020 + \nu_{4B}$

SA — DIODE — KLYSTRON

$\nu_3 = 10.718.068.6$
H$_2$O (28 μm)

$\nu_3 = 12\nu_2 + 0.029 + \nu_{3B}$

SA — DIODE — KLYSTRON

$\nu_2 = 0.890.760.550$
HCN (337 μm)

$\nu_2 = 12\nu_1 + \nu_{2B}$

SA — DIODE

$\nu_1 = 0.074.232.545.83$
KLYSTRON

$\nu_1 = 7\nu_0 + \nu_{1B}$

SA — DIODE

$\nu = 0.010.600.363.69$
KLYSTRON — COUNTER — Cs FREQ STANDARD

LASER FREQUENCY SYNTHESIS CHAIN

(ALL FREQUENCIES IN THz)

⊗ DIODE

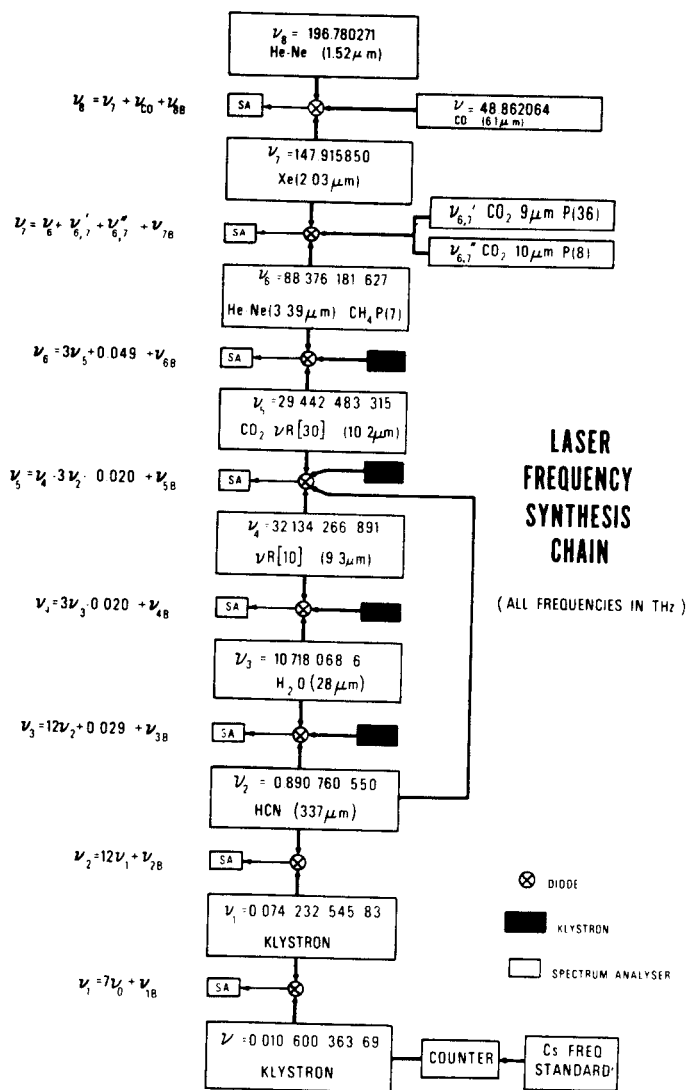▬ KLYSTRON

▢ SPECTRUM ANALYSER

Fig. 4. Laser frequency synthesis chain from Cs (0.009 THz) to 197 THz.

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