

## Direct Frequency Measurement of the 260 THz (1.15 $\mu\text{m}$ ) $^{20}\text{Ne}$ Laser: And Beyond

D.A. Jennings, F.R. Petersen, and K.M. Evenson

Time and Frequency Division, National Bureau of Standards  
Boulder, CO 80303, USA

### ABSTRACT

Absolute frequency measurement has been extended to the visible spectrum with the measurement of the strong 1.15  $\mu\text{m}$  laser line in  $^{20}\text{Ne}$  at 260 THz and lines in iodine at twice this frequency. The 260 THz frequency was synthesized in nonlinear crystals of  $\text{CdGeAs}_2$  and  $\text{AgAsS}_3$  from stabilized  $\text{CO}_2$  lasers and the 1.5  $\mu\text{m}$  laser line in  $^{20}\text{Ne}$ . The visible frequencies were synthesized by generating the second harmonic of the 260 THz radiation with a  $\text{LiNbO}_3$  crystal. The absolute frequencies of ten hyperfine components of  $^{127}\text{I}_2$  near 520 THz were measured.

### INTRODUCTION

Since the frequency measurement by Hocker et al. [1] in 1967 of the 890 GHz line of the HCN laser the progress in laser frequency measurement has been steady (See Fig. 1). The frequency measurement of the 10 THz line of  $\text{H}_2\text{O}$

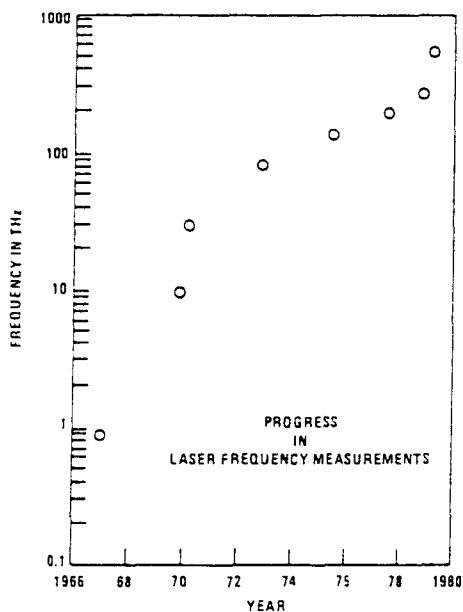


Fig. 1 Progress in laser frequency measurements.

in 1970 by Evenson et al. [2] and the 1973 frequency measurement of the CO<sub>2</sub> laser at 30 THz opened the way for the 88 THz (3.39 μm) frequency measurement of the CH<sub>4</sub> stabilized Ne line by Evenson, et al. [3]

Since the 88 GHz measurement, progress toward higher frequencies has been somewhat slower due to the falloff in the sensitivity of the point-contact MIM diode used in the measurements. Nevertheless, in 1974 the 148 THz frequency of Xe was measured [4], and then, in 1977 the 197 THz Ne radiation was also measured [5]. Efforts to reach 260 THz and the visible were still elusive. During the past year, however, a change in experimental techniques has resulted in attainment of both of these goals.

#### The 260 THz Measurement

In principle, the measurement of the 260 THz (1.15 μm) laser line in <sup>20</sup>Ne is straightforward as can be seen in Fig. 2. The addition of two CO<sub>2</sub> laser lines to the previously measured 197 THz laser radiation of <sup>20</sup>Ne synthesizes the required frequency of 260 THz within the 1.5 GHz pass band of the RF amplifier used.

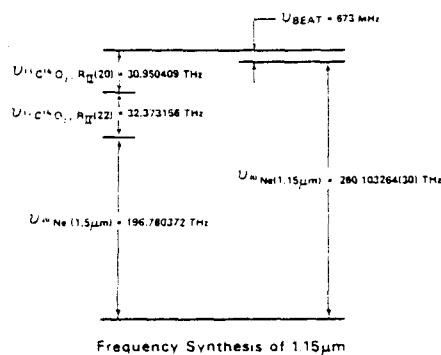


Fig. 2 Synthesis of the 1.15 μm <sup>20</sup>Ne radiation.

The actual synthesis of 260 THz was achieved by using the quadratic nonlinear susceptibility in crystals to mix the known laser frequencies in the following manner. A crystal of CdGeAs<sub>2</sub> was used to sum the R<sub>17</sub>(20), <sup>13</sup>C<sup>16</sup>O<sub>2</sub> and the R<sub>17</sub>(22), <sup>12</sup>C<sup>16</sup>O<sub>2</sub> laser frequencies. The 63 THz output frequency (4.7 μm) was then summed with the 197 THz frequency of a <sup>20</sup>Ne, 1.5 μm laser in a crystal of Ag<sub>3</sub>AsS<sub>3</sub> (proustite). This synthesized radiation (260 THz) was combined with the 260 THz <sup>20</sup>Ne, 1.15 μm laser radiation, and the difference frequency (i.e., the beat frequency) was detected on a fast photovoltaic Ge diode. The resulting beat frequency was amplified and measured with a spectrum analyzer.

The experimental setup is shown in Fig. 3. The CO<sub>2</sub> reference lasers were stabilized to the saturated absorption in CO<sub>2</sub>. [5] and the CO<sub>2</sub> power lasers were frequency offset locked. The 1.15 μm, <sup>3</sup>He-<sup>20</sup>Ne laser was frequency offset locked to a Lamb-dip stabilized 1.15 μm, <sup>20</sup>Ne laser [6]. The 1.5 μm, <sup>3</sup>He-<sup>20</sup>Ne laser was manually set to the center of its gain curve. The basic lasers have been described elsewhere with the exception that mirrors and gas fills were changed to enhance laser performance for each particular frequency [3,7].

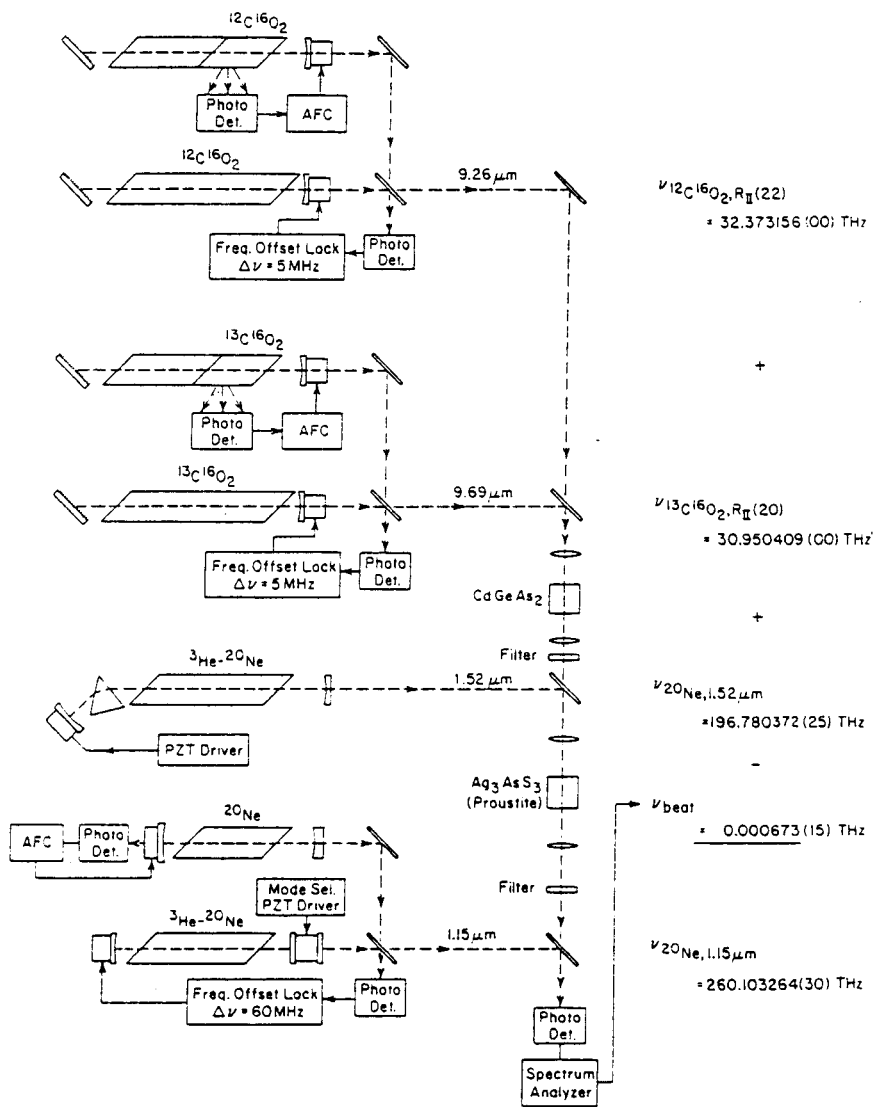


Fig. 3 A block diagram of the 260 THz experimental setup with the final result.

Therefore, the frequency of the  $^{20}\text{Ne}$ , 1.15  $\mu\text{m}$  laser was:

$$\begin{aligned} \nu_{^{20}\text{Ne}, 1.15 \mu\text{m}} &= \nu_{^{12}\text{C}^{16}\text{O}_2, R_{II}(22)} + \nu_{^{13}\text{C}^{12}\text{O}_2, R_{II}(20)} \\ &\quad + \nu_{^{20}\text{Ne}, 1.52 \mu\text{m}} - \nu_{\text{beat}}, \end{aligned}$$

where

$$\nu_{^{12}\text{C}^{16}\text{O}_2, R_{II}(22)} = 32.373\,156(00) \text{ THz [8]},$$

$$\nu_{^{13}\text{C}^{16}\text{O}_2, R_{II}(20)} = 30.950\,409(00) \text{ THz [9]},$$

$$\nu_{^{20}\text{Ne}, 1.52 \mu\text{m}} = 196.780\,372(25) \text{ THz [10]}, \text{ and}$$

$$\nu_{\text{beat}} = 0.000\,673(15) \text{ THz.}$$

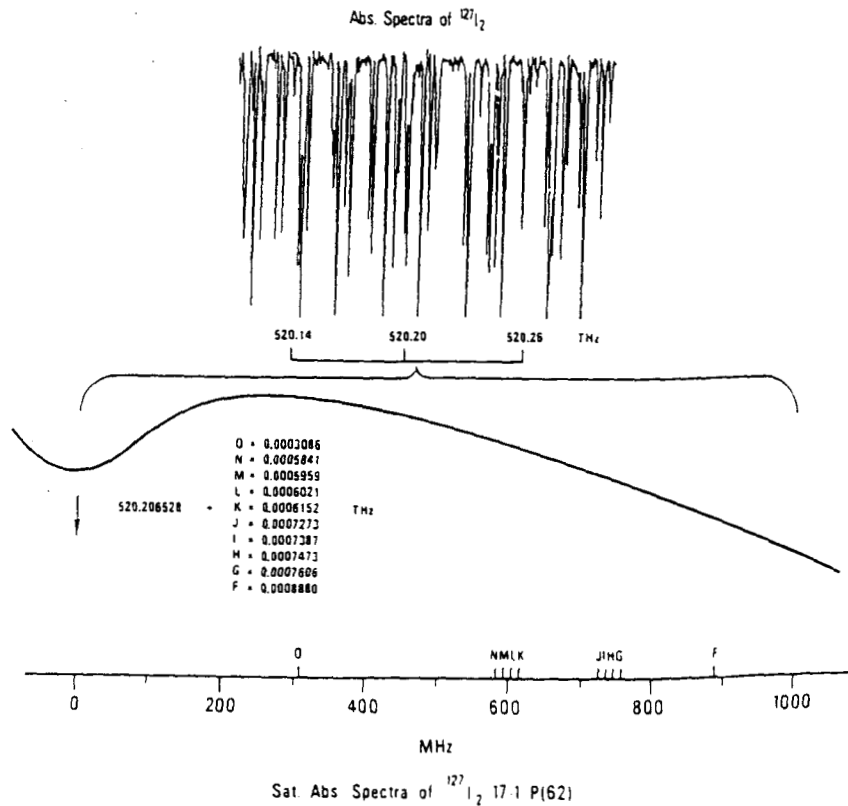
Thus,

$$\nu_{^{20}\text{Ne}, 1.15 \mu\text{m}} = 260.103\,264(30) \text{ THz.}$$

This number is in agreement with the frequencies derived from wavelength measurements in the spectra tables [11] and a recent, more accurate wavelength measurement [12]. The uncertainty in the  $^{20}\text{Ne}$ , 1.15  $\mu\text{m}$  frequency comes from the uncertainties in the  $^{20}\text{Ne}$ , 1.52  $\mu\text{m}$  frequency (25 MHz) and the determination of line center (15 MHz).

Previous frequency measurements up to 197 THz (1.52  $\mu\text{m}$ ) have utilized the tungsten-nickel, point-contact diode as the nonlinear element for synthesis and detection. Several unsuccessful attempts were made to use this diode in the measurement of the 260 THz frequency prior to the measurement described above. After this frequency had been successfully measured with the nonlinear crystals (i.e. the beat frequency was known) the 4.73, 1.52, and 1.15  $\mu\text{m}$  radiations were again focused on the point-contact diode in another attempt to use this device. All rectified signals were of the order of 1 mV; the polarity of the 1.52 and 1.15  $\mu\text{m}$  signals was opposite to that of the 4.73  $\mu\text{m}$  signal. A search was made at the known beat frequency with diode impedances from a few hundred ohms to several thousand ohms. The results were unsuccessful.

Once the 260 THz  $^{20}\text{Ne}$  line is measured it is in principle a simple matter to double this frequency and thereby synthesize a known frequency in the visible, 520 THz. In a joint experiment with the National Research Council in Ottawa, Canada ten hyperfine transitions in  $^{127}\text{I}_2$  near 520 THz were measured by comparison with the known frequency of the Lamb-dip stabilized pure  $^{20}\text{Ne}$  laser at 260 THz. The yellow-green light at 520 THz, generated (in the NRC laser) [13] by intracavity doubling in lithium niobate of 260 THz radiation from a He-Ne discharge, was servo-locked to individual hyperfine components of  $^{127}\text{I}_2$  observed in saturated absorption, and their frequencies were determined simply by measurement of the beat frequencies of the two radiations at 260 THz.



at the 2<sup>nd</sup> Harmonic of the  $^{20}\text{Ne}$  260 THz Laser

Fig. 4 Results of the  $^{127}\text{I}_2$  hyperfine frequency measurement at 520 Hz. Absorption spectra of  $^{127}\text{I}_2$  from the work of GERSTENKORN et al. [16]

There is a substantial overlap of the doubled radiation and the strong P(62) line in the 17-1 band of  $^{127}\text{I}_2$  (See Fig. 4). Fifteen hyperfine components (labeled a to o in order of decreasing frequency) are expected in this line, and ten of these (f to o) were observed in saturated absorption within the laser tuning range. The lowest frequency component, o, is well separated from the others and provides by far the best signal-to-noise ratio because the background absorption from the other iodine components is relatively small and because it occurs near the peak of the laser output power. The components had a full width at half height of 2 MHz for an iodine pressure of 4 Pa. With 2 MHz frequency modulation (at 1.8 kHz), the laser could be servo-locked to the zero-crossing of the amplitude modulation at 5.4 kHz that occurs at the center of each component (third harmonic locking). The infrared laser was locked in turn to each hyperfine component for the frequency measurements described below, and thus was at half the frequency of the hyperfine line.

The frequency measurements were done simply by combining the 260 THz beams from the two lasers on a high speed photodiode. The beat frequency

was displayed on a spectrum analyzer and measured with an adjustable marker oscillator and counter. The frequency,  $f_\alpha$ , of the  $\alpha$ -component is given by

$$f_\alpha = 2 [f_{\text{Ne}} + (f_o/2 - f_{\text{Ne}})].$$

Six determinations of the frequency difference between the o-component and the Lamb-dip were made with a readjustment of the mirrors of the NBS laser for symmetrical Lamb-dip between each one. The standard deviation of six such settings was about 0.1 MHz. Systematic errors due to asymmetry of the modulation envelope were estimated to be less than about 1% of the 45 MHz full width of the Lamb-dip. Other sources of error were significantly less than this. The mean value is

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

and the estimated 1- $\sigma$  error is 0.5 MHz. The frequency of the o-component of P(62), 17-1 band of  $^{127}\text{I}_2$  is thus

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

A preliminary measurement of the wavelength of this component gave  $\lambda = 576\,294\,758 \pm 6 \text{ fm}$  [14], from which we calculate  $f_o = c/\lambda = 520\,206\,811 \pm 6 \text{ MHz}$  [15]. The agreement between the above values for the frequency is satisfactorily within the error limits. In addition, the laser was locked to each of the components ( $n$  to  $f$ ) and the beat frequencies between  $f_o/2$  and  $f_{\text{Ne}}$  were measured. The results are shown in Fig. 4. The uncertainty in each of these beat frequencies is also 0.5 MHz.

This extension of absolute frequency measurements to the visible paves the way for highly accurate measurements in this portion of the electromagnetic spectrum. The rather large error limit on  $f_\alpha$  is due to the free running 197 THz He-Ne laser used in the measurement of the Lamb-dip stabilized  $^{20}\text{Ne}$ , 1.15  $\mu\text{m}$  laser. In view of the reproducibility of this Lamb-dip stabilized laser, an improved determination of its frequency,  $f_{\text{Ne}}$ , can be combined with the above value of  $f_o/2 - f_{\text{Ne}}$  to decrease the uncertainty of these iodine frequencies by about two orders of magnitude.

#### The Frequency Chain to the Visible

Fig. 5 illustrates the entire chain of frequency measurements which link the frequency at 520 THz to the Cs frequency standard. 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. Unfortunately, such a stabilization technique does not exist for the 2.03  $\mu\text{m}$  Xe and 1.52  $\mu\text{m}$  Ne lasers. Consequently, the principal uncertainty in the 520 THz measurement results from the uncertainties in these two frequencies. With this chain, a significant improvement can therefore be realized by connecting the 88 THz and 260 THz lasers in a single step. This measurement can be done in a straightforward way by simultaneously observing beats between the 88 and 148 THz lasers, the 148 and 197 THz lasers, and the 197 and 260 THz lasers. Unfortunately, this measurement would require eleven lasers and one klystron in a major experimental effort.

Therefore, the chain shown in Fig. 6 is proposed as an alternative technique for connecting the  $^{127}\text{I}_2$  stabilized 520 laser to the Cs frequency standard. The multiplication factor of 48020 is accomplished with six lasers and five klystrons, a significant reduction from the previous chain.

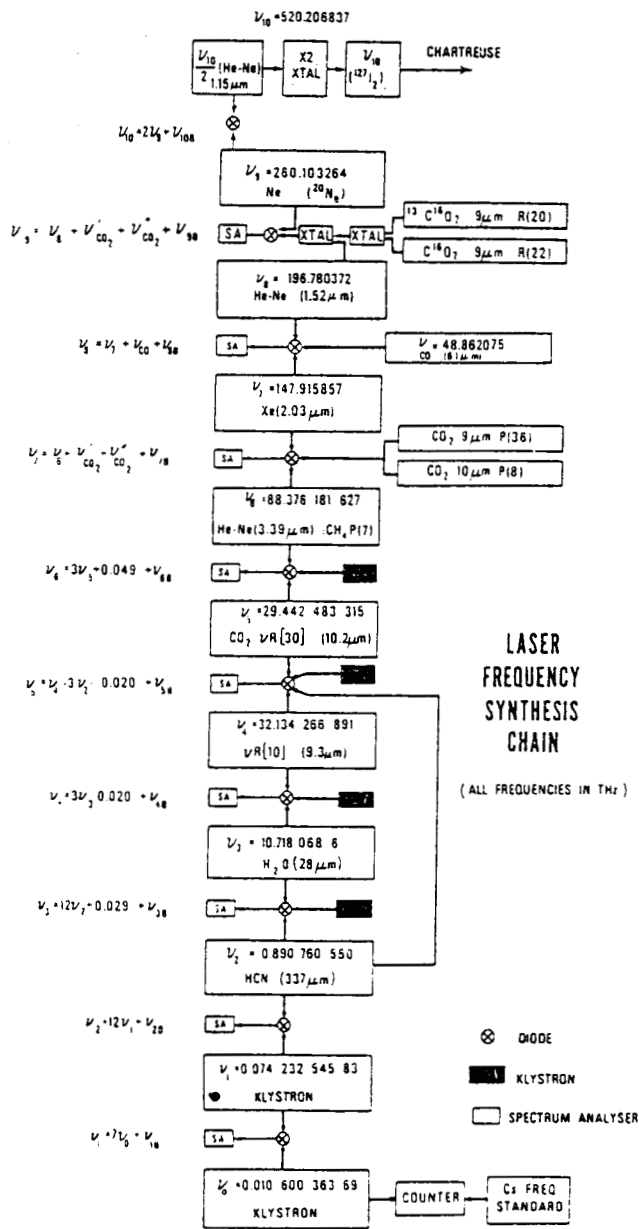


Fig. 5 Old laser frequency synthesis chain from X-band to visible.

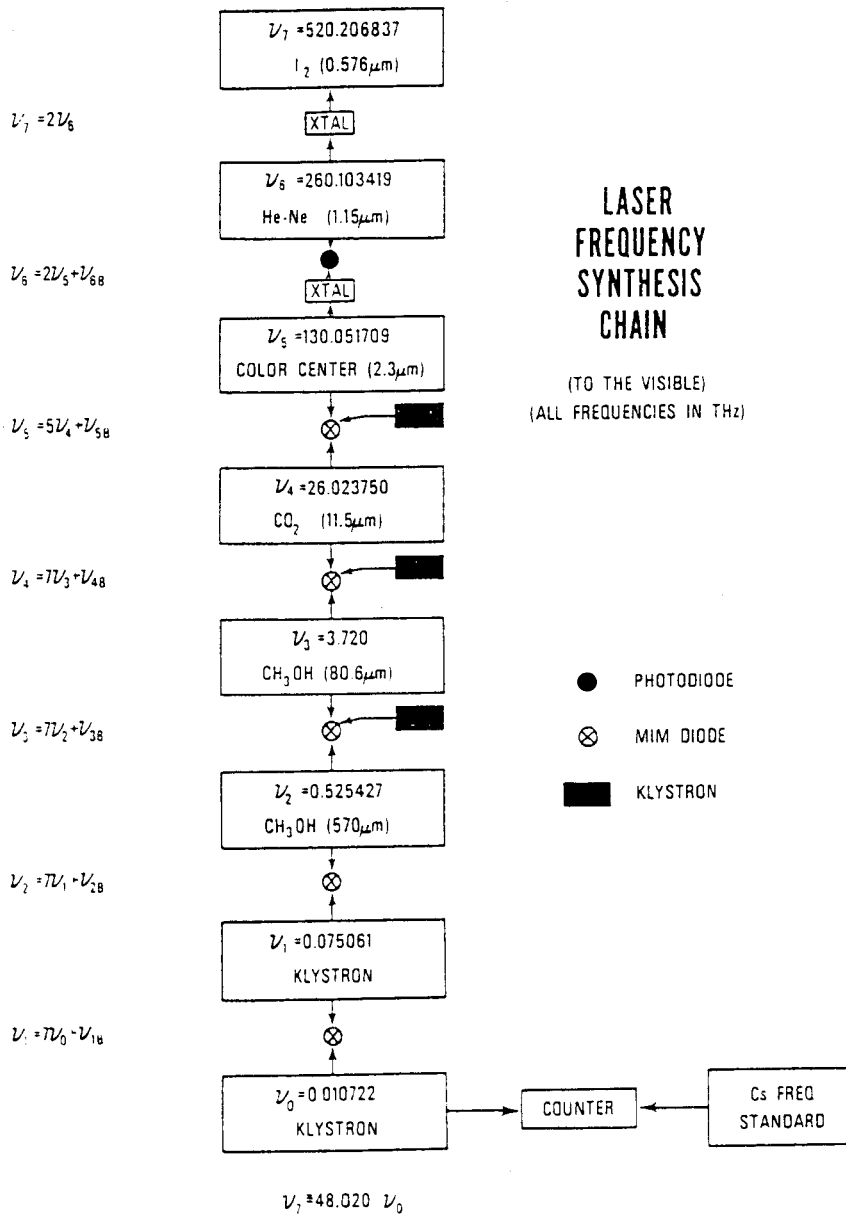
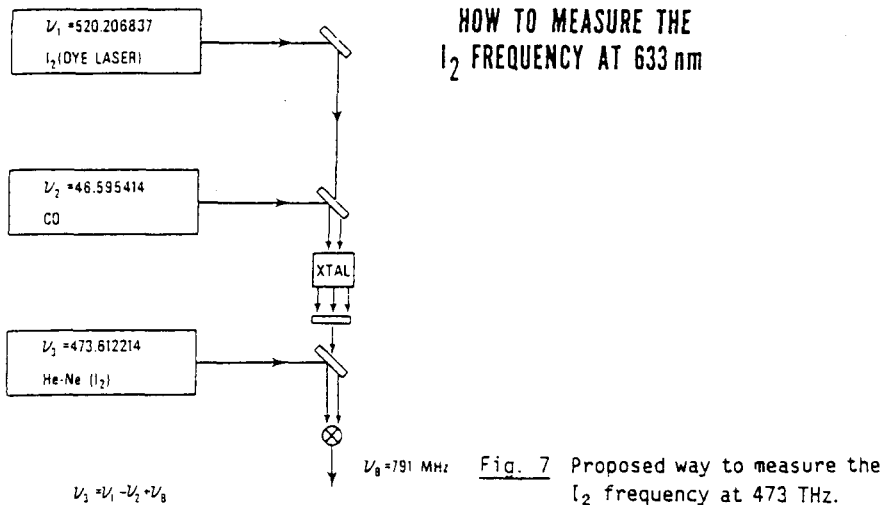


Fig. 6 New proposed simplified laser frequency synthesis chain.



## HOW TO MEASURE THE I<sub>2</sub> FREQUENCY AT 633 nm



All lasers in the chain exist, and each multiplication step has been demonstrated to be experimentally feasible with the nonlinear devices indicated in the diagram. This chain should have a measurement capability with a fractional frequency uncertainty between  $10^{-10}$  and  $10^{-11}$  for the I<sub>2</sub> transition at 520 THz.

This stabilized frequency at 520 THz appears to be an ideal reference from which to synthesize other standard frequencies in the visible spectrum. It is anticipated that these frequencies will again be synthesized by mixing the radiation from the 520 THz laser with an appropriate infrared laser in a nonlinear crystal such as AgGaS<sub>2</sub>. This crystal has been produced with a wide, low absorption, transmission band ( $\alpha < 0.5 \text{ cm}^{-1}$  for  $0.5 < \lambda < 10 \text{ } \mu\text{m}$ ) [16] and has been used to up-convert  $10.6 \text{ } \mu\text{m}$  radiation into the green spectral range [17]. As an example of this method, Fig. 7 shows a possible way to measure the frequency of the I<sub>2</sub> stabilized  $0.633 \text{ } \mu\text{m}$  Ne laser.

### Summary

In summary, we have demonstrated the technique of visible frequency measurements that are directly related to the CS frequency standard. In the future we would hope to use this technique to generate a number of standard frequencies in the visible.

### References

1. L. O. Hocker, A. Javan, and D. Ramachandra Rao, Appl. Phys. Lett. 10, 147 (1967).
2. K. M. Evenson, J. S. Wells, L. M. Matarrese, and L. B. Elwell, Appl. Phys. Lett. 16, 159 (1970).
3. K. M. Evenson, J. S. Wells, F. R. Petersen, B. L. Danielson, and G. W. Day, Appl. Phys. Lett. 22, 192 (1973).

4. D. A. Jennings, F. R. Petersen, and K. M. Evenson, *Appl. Phys. Lett.* 26, 510 (1975).
5. K. M. Evenson, D. A. Jennings, F. R. Petersen, and J. S. Wells, *Laser Spectroscopy III*, J. L. Hall and J. L. Carlsten, eds. (Springer-Verlag, Berlin 1977), pp. 56-68.
6. J. H. Hall, *IEEE, J. Quantum Electron.*, QE4, 638 (1968).
7. K. M. Evenson, J. S. Wells, F. R. Petersen, B. L. Danielson, G. W. Day, R. L. Barger, and J. L. Hall, *Phys. Rev. Lett.* 29, 1346 (1972).
8. F. R. Petersen, D. G. McDonald, J. D. Cupp, and B. L. Danielson, "Laser Spectroscopy", edited by R. G. Brewer and A. Mooradian (Plenum, New York, 1974), pp. 171-191.
9. Charles Freed, A. H. M. Ross, and Robert G. O'Donnell, *J. Mol. Spectrosc.* 49, 439 (1974). Quoted error is a result of recent measurements with stabilized lasers (saturated absorption). Private communication with Charles Freed.
10. The frequency of the 1.52  $\mu\text{m}$  He-Ne laser was remeasured and the corrected value is listed. In the previous measurement (K. M. Evenson, D. A. Jennings, F. R. Petersen, and J. S. Wells, in *Laser Spectroscopy III*, J. S. Hall and J. L. Carlsten, eds. (Springer-Verlag, Berlin 1977), pp. 56-68.), a higher frequency mode of the 1.52  $\mu\text{m}$  laser was not detected, the CO frequencies were not directly measured, and the 1.5  $\mu\text{m}$  laser discharge tube was filled to a total pressure of 324 Pa (with He:Ne=10:1) instead of the 648 Pa used in the present experiment. The new measurements, made with the  $^{12}\text{C}^{16}\text{O}$ ,  $P_{18}(14)$  line only, revealed the following corrections: (1) Mode +71 MHz; (2) directly measured frequency of  $^{12}\text{C}^{16}\text{O}$ ,  $P_{18}(14)$  [ $^{12}\text{C}^{16}\text{O}$ ,  $P_{18}(14)$  = 48.862 075(3) THz] + 11 MHz; and (3) pressure shift [+65(9)kHz/Pa] + 21 MHz. The resultant frequency is  $^{20}\text{Ne}$ , 1.5  $\mu\text{m}$  = 196.780 372(25) THz.
11. Charlotte E. Moore, "Atomic Energy Levels", Vol. I, NSRDS-NBS35, National Bureau of Standards, 1971 (U.S. GPO, Washington, D.C. 20402).
12. The wavelength of 1.15  $\mu\text{m}$  is from J. L. Hall and S. A. Lee, private communication, using the techniques of J. L. Hall and S. A. Lee, *Appl. Phys. Lett.* 29, 367 (1976).
13. G. R. Hanes, N.R.C. Ottawa, Canada, paper in preparation.
14. The authors wish to thank Dr. K. H. Hart of NRC, Ottawa, for this wavelength value.
15. Using the value  $c = 299\,792\,458$  M/s recommended in BIPM, *Comptes Rendus des Séances de la Conf. Gen. des Poids et Mesures*, 15th, p. 103, 1975.
16. S. Gerstenkorn and P. Luc, "Atlas du Spectre d'Absorption de la Molécule d'Iode," Centre National de la Recherche Scientifique, Paris, 1978.
17. H. Matthes, R. Viehman, and N. Marschall, *Appl. Phys. Lett.* 26, 237 (1975).
18. Tien-Lac Hwang and S. E. Schwarz, *Appl. Phys. Lett.* 31, 99 (1977).