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OPTICAL FREQUENCY MEASUREMENT AT NRC AND PROGRESS TOWARDS FREQUENCY MEASUREMENT OF VISIBLE LIGHT

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Abstract.- This article describes the frequency comparison chain that is being set up at N.R.C. (Ottawa) with the aim of making possible frequency measurement with respect to the Cs primary standard, of standards throughout the spectrum up to the visible region. Progress to date includes demonstration of phase locking of several stages up to 30 THz, demonstration of the feasibility of several stages from 30 THz to 260 THz and operation of the last stage from 260 THz to 520 THz (0.576 μm).

Introduction.- In order to realize the use of the same standard for length and time, a frequency chain is required to relate microwave to optical frequencies. This is so not only for the process of establishing the new definition of length in terms of the Cs 9 GHz transition, as now proposed, but also in the likely event that a new standard will replace the Cs standard in the not too distant future. An improved new standard might well be provided by the sharp resonances in OsO_4 or SF_6 at about 30 THz (10 μm) or some other of the proposals discussed in this conference; the need for such a chain is also implicit in the increasing use by spectroscopists of standards throughout the infrared, based on frequency measurements.

The frequency chain being set up at N.R.C. in Ottawa, to fill this need, is conveniently described as two parts: the first, operating in the region where point contact diodes operate as harmonic generators, aims at producing CO_2 laser emission in the 10 μm (30 THz) region, which will be phase locked to the Cs primary standard; it is intended for use not only as a base to extend frequency measurements to the visible spectrum, but also to compare the frequencies and reproducibilities of the Cs standard and the above mentioned lines in the 30 THz region. The second part, covering the region through which the electrical response of point contact diodes appears to fall off and non linear optical crystals must be used, aims at extending the chain to standards in the visible spectrum.

Microwave to 30 THz Chain.- The microwave to 30 THz part of the chain is being set up by B.G. Whitford. It makes use of intermediate frequencies generated in the diode by simultaneous radiation from two CO_2 lasers, operating on appropriately different transitions, instead of using frequencies directly generated by materials such as HCN, H_2O , CH_3OH etc., as has been the practice in other laboratories. This method has the advantage of using only simple, convenient CO_2 lasers, although this advantage is somewhat offset by the relatively weak signals provided by the difference frequencies resulting in the requirement of an extra step.

The chain has already been operated in a basic form for making a preliminary measurement of the speed of light by measurement of the frequency of a CH_4 stabilized He-Ne laser, as previously described (1,2); figure 1 shows one of several alternative schemes: the output of a .071 THz klystron,

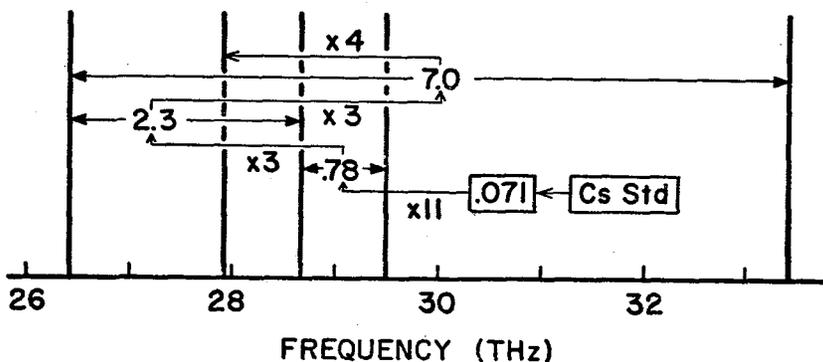


Fig. 1 Frequency chain to 28 THz using differences between CO_2 laser lines.

locked to the Cs standard, is radiated onto a diode where its 11th harmonic beats with a difference frequency of .78 THz, generated in the same diode by simultaneous irradiation from two carbon dioxide lasers operating on the transitions $9 \mu\text{m R}(30)$ of $^{12}\text{C}^{16}\text{O}_2$ and $9 \mu\text{m P}(6)$ of $^{12}\text{C}^{16}\text{O}_2$ respectively. Similarly, the third harmonic of the above difference frequency is generated and mixed in a second diode with a 2.3 THz difference frequency generated from the transitions $9 \mu\text{m P}(6)$ of $^{12}\text{C}^{16}\text{O}_2$ and $10 \mu\text{m P}(40)$ of $^{13}\text{C}^{16}\text{O}_2$. In the third step the third harmonic of the 2.3 THz difference is mixed with a 7.0 THz difference frequency generated from the transitions $10 \mu\text{m P}(40)$ of $^{13}\text{C}^{16}\text{O}_2$ and $9 \mu\text{m R}(48)$ of $^{12}\text{C}^{18}\text{O}_2$ respectively. Finally the fourth harmonic of the 7.0 THz difference is mixed with the 28 THz signal from the single laser operating on the $10 \mu\text{m R}(6)$ line of $^{13}\text{C}^{18}\text{O}_2$. In each stage the beat frequencies (at approximately 729, 592, 1431 and 2611 MHz respectively) were measured against the frequency standard, thus giving the frequency of the 28 THz line.

In the above chain, each stage was operated and measured as a separate experiment, with the result that the final accuracy was limited to about two parts in 10^9 , because of the relatively large proportional errors in the differences between lasers that were independently stabilized (by saturated fluorescence) and because of the multiplying effect of the harmonic factors. This error effect ought largely to disappear when the intermediate stages are used as servo-controlled idlers in the complete phase-locked chain that is now being set up.

In the newly constructed chain, the five lasers are mounted on a heavy, vibration isolated, concrete table inside a concrete block housing (approximately 4 m long x $2 \frac{1}{2}$ m wide x $2 \frac{1}{2}$ m high) which serves to give protection from airborne vibrations and temperature effects. The resultant free running stability of each laser (about 2 KHz width in 1 second observation time) is much improved over the former setup. The phase-locking servo control is now being developed and two important results have so far been achieved. In the first, two lasers were phase-locked, offset exactly 1340 MHz (referred to the Rb standard), using a signal deliberately attenuated to a value comparable to the most difficult stage of the above described chain; this success showed: that the W-Ni diode is a suitable mixer for such purposes, that U.H.F. locking is possible with -110 dbm signals, that locking requires only a commercial piezo translator, and that only one servoloop is required. In the second result, the third harmonic of a difference frequency was beat against

another difference frequency (stage two of the chain of figure 1) and phase-locking was attempted, but the imperfectly optimized servoloop would only lock for a fraction of a second, a fault that is now being corrected; the free running beat signal is shown in figure 2. These two results have shown that the complete phase-locked chain has an excellent chance of operating

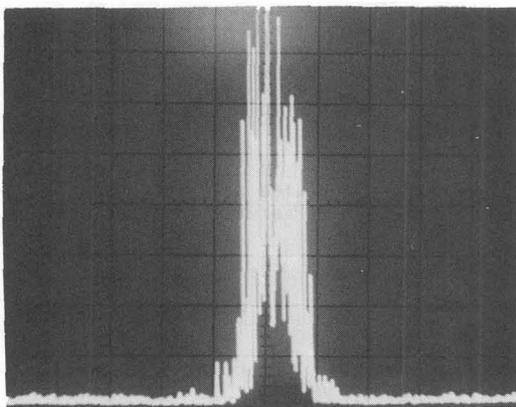


Fig. 2 Free running 20 MHz beat from mixing $615 - [(f_2 - f_3) - 3(f_1 - f_2)] = 20 \text{ MHz}$. Hor. 5 kHz/div., scan 200 ms/div.

successfully when a number of improvements in the electronics have been made.

It can be argued that the measurement of the 30 THz frequencies could be made, without the difficulty of phase-locking, by the use of simultaneous, directional counting of the beats at each stage during the measurement. Although the difficulties of phase-locking such small signals are not trivial, it appears evident to us that the system will work and therefore is the preferred method; certainly it would make a much more sanitary and informative method for comparing the frequencies and characteristics of the SF_6 and OsO_4 resonances with the Cs standard, if one could compare two signals that differ by a few megahertz, each locked to one of the standards being compared. Amplification of phase jitter will not present the usual problems because most of the stages can be servo-controlled to follow the upper frequency at each step.

It is worth noting that, in addition to the above special application of frequencies synthesized from the differences between transitions in the CO_2 9 μm & 10 μm bands, the technique can be used to measure frequencies ranging from a few megahertz to about seven terahertz, or frequencies differing by such amounts from known standards, within the range over which the point contact diodes operate electrically. The lines of the normal bands of CO_2 lines can be supplemented by the very large number of lines provided by the sequence bands, by the use of unusual isotopic composition, etc. An interesting example of the technique is the method that has been used by K. Siemsen at N.R.C. to measure the frequency of a laser well outside the grid of CO_2 reference lines. As shown in fig. 3, when two appropriate known frequencies f_1 and f_2 are mixed with the unknown frequency f_3 , the latter can be deduced from the observed beat frequency $f_b = (f_1 - f_2) - (f_2 - f_3)$.

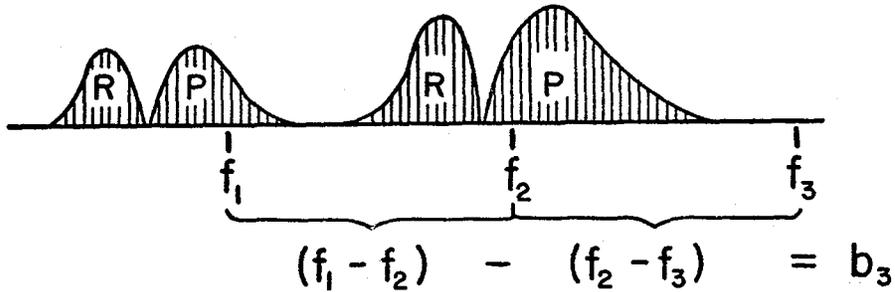


Fig. 3 Measurement of frequency f_3 outside reference band by observing beat b_3 between difference frequencies.

Frequency Chain 30 THz to the Visible.- Frequency comparison up to 88 THz, by using point contact diodes to generate and mix harmonics of 30 THz lines, is relatively straightforward; it has been done in at least five laboratories, resulting in the well confirmed value for the speed of light proposed for a new definition of the Metre (3). Above this range, the electrical response of the diode appears to fall off rapidly and only few applications have been made, notably those of Evenson and his colleagues at N.B.S. (Boulder) who succeeded in measuring several lines up to 196 THz, including the Xe 3.5 μm , Xe 2.02 μm and Ne 1.5 μm laser lines (4,5). These measurements were made by using sums of frequencies (not harmonics) and attempts to go higher with point contact diodes have so far failed. The experiments of Walther and his colleagues at Garching (6) and Meisel at Bonn (7) to measure large frequency differences in the visible by point contact and Schottky diodes involve a different mechanism and the diodes cannot be used as harmonic generators or electrical mixers in the way the point contact diodes are used up to about 200 THz.

In the region above 1.5 μm , it is necessary to use optically non-linear crystals for production of second harmonics and for mixing frequencies. However a frequency chain in this region is far from easy because of the large frequency differences between the relatively few suitable laser lines available, coupled with the problem of finding crystals that satisfy the conditions of transparency and phase matching. Figure 4, showing the spectrum on a linear scale, illustrates this problem: the whole range of the frequency chain up to 30 THz is less than the difference between the 1.5 μm and 1.15 μm lines of He-Ne; the width of the frequency scale markers on the graph represents about 10 GHz - well beyond the capability of the usual detectors available for this part of the spectrum. The chance of finding a suitable laser whose harmonic is "within reach" of a suitable standard is very small; and it is a rare crystal that is transparent from 30 THz to the visible and has suitable constants for phase matching to make possible the use of the CO_2 lines for measuring frequency differences between visible lines.

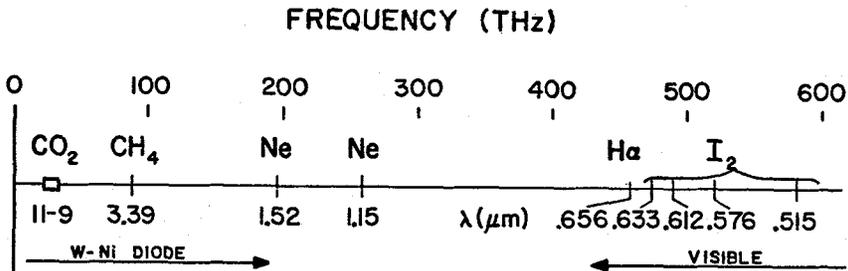


Fig. 4 Linear scale of frequencies to visible region.

The methods proposed by most laboratories, to alleviate these difficulties, make use of tunable color centre and dye lasers, in order to overcome the lack of convenient coincidences of available laser lines with harmonics of other lines. For example, at N.B.S. (Boulder) it is proposed to use a color centre laser that can be tuned to be one half the frequency of the Ne 1.15 μm line which is also approximately five times a known CO_2 transition. The problems of control and stability of such lasers are not trivial however. We have chosen to try to make use of some coincidences that do exist, in order to have certain technical advantages provided by commonly used lasers.

A chain now being tried is shown schematically in figure 5: a He-Xe laser emission

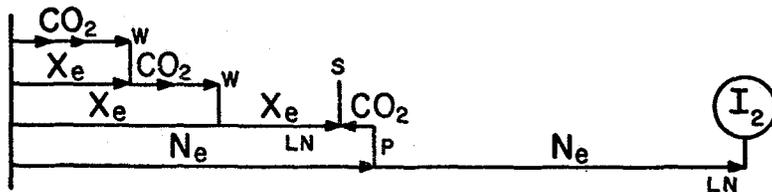


Fig. 5 Chain to measure I_2 line by use of CO_2 , Xe and Ne lines mixed on Schottky²(S) and W-Ni (W) diodes and crystals of proustite (P) and LiNbO_3 (LN).

at 3.5 μm is beat against the sum of three CO_2 lines in a W-Ni diode, as shown; the same-line, added to two other CO_2 lines is used to measure the frequency of a second He-Xe laser line at 2.02 μm , also in a W-Ni diode. The two Xe lines can be excited simultaneously in a single plasma tube and are added by means of an

in-cavity LiNbO_3 crystal to synthesize a line at $1.28 \mu\text{m}$ (233 THz). A second $1.28 \mu\text{m}$ signal is synthesized from the difference frequency between a $1.15 \mu\text{m}$ He-Ne laser and the $10 \mu\text{m}$ P(26) $^{13}\text{C}^{16}\text{O}_2$ laser line by mixing in a proustite crystal. According to known values for the Xe, Ne and CO_2 lines, the two $1.28 \mu\text{m}$ signals are expected to be within about 1 GHz, a difference that can be measured on a Ge Schottky diode. This would yield the frequency of the Ne $1.15 \mu\text{m}$ line in terms of the CO_2 lines, which in turn can be related to the chain described previously. The final stage to the visible is by doubling the $1.15 \mu\text{m}$ Ne line in a LiNbO_3 crystal; it was achieved two years ago (8) and will be reviewed briefly later.

The most difficult part of this chain, that connecting the Xe and Ne lines, is being set up by H. Riccius and D. Smith; the relationship of the lasers and mixing crystals is shown schematically in figure 6. The boxes

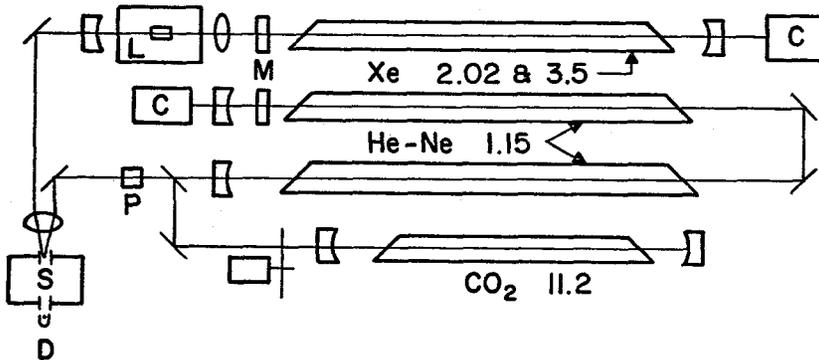


Fig. 6 Schematic for measurement of frequency of Ne 1.15μ line using Xe and CO_2 lasers.

marked "C" represent the frequency stabilizing control, which will eventually be by reference to CO_2 lines for the Xe laser lines and by reference to an I_2 line, as described below, for the Ne laser line. The LiNbO_3 crystal is maintained at $494^\circ\text{C} \pm 0.1^\circ$ in an oxygen atmosphere, for phase matching the $2.02 \mu\text{m}$, $3.5 \mu\text{m}$ and $1.28 \mu\text{m}$ lines.

The progress to date in operating this part of the chain is as follows: The Xe lines, emitted by one laser operating simultaneously on two transitions, have been mixed in the LiNbO_3 crystal to produce about $1 \mu\text{W}$ of power at $1.28 \mu\text{m}$ with a $\text{S/N} \approx 25 \text{ db}$ in about 1 second averaging time. A signal of about the same power and S/N was generated from He-Ne and CO_2 lines having powers of about 50 mW and 2 watts respectively.

The sum generation from the two Xe lines was relatively straightforward, except for difficulties associated with the rather high phase matching temperature and the very accurate control required. The mixing of the Ne and CO_2 lines in the proustite presented problems because, at wavelengths as long as the required CO_2 transition ($11.2 \mu\text{m}$), absorption is appreciable. As a result, if sufficient power for the difference frequency generation is applied continuously, the crystal overheats and is damaged or at least is unstable. This problem was overcome by chopping the CO_2 beam at 77 Hz in such a way that the beam was off for 90% of the time, during which time the detection circuitry was also gated off; under these conditions the CO_2 power could be as high as 10 W without damage to the crystal. According to the literature (9), type II

phase matching ought to produce significantly less absorption at $11.2 \mu\text{m}$ at a sacrifice of only a factor two in the conversion efficiency. However, it was in fact found that heating effects appeared to be worse with type II matching and so type I will be used.

A search for the beat between the two synthesized $1.28 \mu\text{m}$ signals is now in progress. Although the individual signals obtained give a large S/N with one second averaging, the jitter in the 1 GHz beat due to laser instabilities will make necessary a much greater bandwidth for the search and it may therefore be difficult to pick up the beat signal without more effective means for preliminary stabilization than installed at present; once the beat is found, stabilization of one of the $1.28 \mu\text{m}$ signals with respect to the other ought not be too difficult.

An alternative to the above method of relating the $1.15 \mu\text{m}$ line to the CO_2 lines that is being considered is illustrated schematically in fig. 7. This system would utilize a W-Ni diode to compare directly a

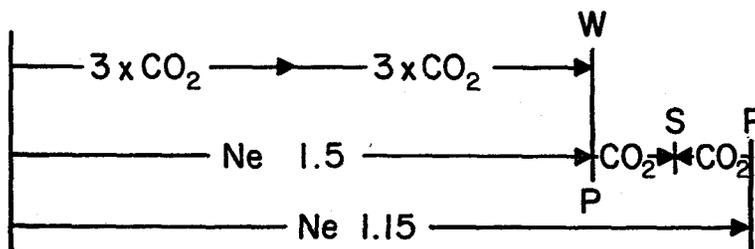


Fig. 7 Chain to measure frequency of Ne $1.15 \mu\text{m}$ line using Ne $1.5 \mu\text{m}$ and CO_2 .

$1.5 \mu\text{m}$ He-Ne laser output to the sum of the third harmonic of the $9 \mu\text{m}$ R(18) line of $^{12}\text{C}^{18}\text{O}_2$, and the third harmonic of the $9 \mu\text{m}$ R(44) line of $^{12}\text{C}^{16}\text{O}_2$. The $1.5 \mu\text{m}$ line would then be added to the $9 \mu\text{m}$ R(22) $^{12}\text{C}^{16}\text{O}_2$ line and compared to the difference between the $1.15 \mu\text{m}$ line of Ne and the $9 \mu\text{m}$ R(20) $^{13}\text{C}^{16}\text{O}_2$ line, both syntheses being made in proustite. Unlike the case in the previously described scheme, the proustite is used in a region ($9 \mu\text{m}$) where absorption is low. The main uncertainty is whether the signal for the first step can be seen. Evenson (Priv. Comm.) has tried to beat the $1.5 \mu\text{m}$ line against the sixth harmonic of a CO_2 line, plus a klystron frequency, but was unsuccessful in finding a signal. We are nevertheless encouraged by the fact that we are using a harmonic order of one less (the klystron frequency is not required) and Evenson has shown that the diode apparently responds electrically in this region. Success in this step may simply be a question of using sufficiently well stabilized lasers to realize a much narrower bandwidth.

The last stage in this part of the chain set up by G. Hanes was achieved at N.R.C. in 1979 (8) by doubling the He-Ne $1.15 \mu\text{m}$ laser emission and comparing the doubled radiation to I_2 absorption lines, as shown in fig. 8. It employed a doubly resonant cavity indicated by the paths ABCBE

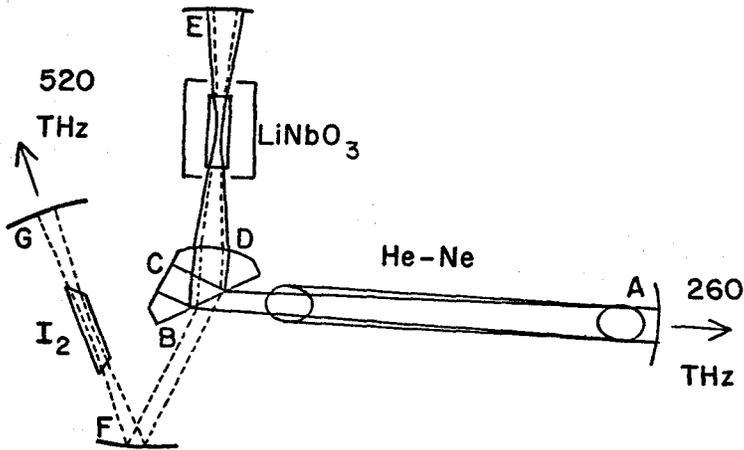


Fig. 8 Schematic for stabilizing doubled He Ne $1.15 \mu\text{m}$ line by reference to I_2 line at $.576 \mu\text{m}$.

and EFBG. The $1.15 \mu\text{m}$ radiation produced by the He-Ne plasma tube is focussed into the LiNbO_3 crystal which is accurately temperature controlled (at 173.0°C), for phase matching to produce the doubled frequency at $.576 \mu\text{m}$ (520 THz). An additional phase matching requirement is satisfied by the special dispersive reflector at E to ensure that the reflected second harmonic is in phase with the second harmonic generated by the reflected fundamental. An I_2 cell in the second cavity, resonant at the doubled frequency, produces saturated absorption features that can be used for stabilization. Scanning and servo-control of the cavity arms AC, CE and EG are suitably coordinated so as to be resonant simultaneously at the required frequencies. About $100 \mu\text{W}$ of $1.15 \mu\text{m}$ radiation was emitted at A and about $20 \mu\text{W}$ of $0.576 \mu\text{m}$ radiation at G, both being frequency controlled by the same I_2 absorption line. The fortunate I_2 hyperfine spectrum at 520 THz is shown in figure 9; the strong component at the left, where the gain has been reduced by a factor 10, is particularly noteworthy. The line, having a demonstrated Q of at least 3×10^8 , ought to provide a good standard, but a detailed study of its characteristics has not yet been made.

General Remarks.— The frequency chain described above now appears to have an excellent chance of fulfilling the aim of making possible frequency measurement up to the visible part of the spectrum, in terms of the primary standard. However, in order to attain an accuracy that is limited by the reproducibilities of the standards at each end of the chain, i.e. of the order of one part in 10^{13} , a considerable amount of work remains to be done to overcome some knotty technical problems, for this as well as for other proposed chains described in the literature. Although feasibility of the measurement of a visible line has, in a sense, been demonstrated (10), that measurement (of an I_2 line at $.576 \mu\text{m}$) depended on the use of the molecular constants of CO for reference to the primary standard; the most accurate value for the frequency of the line is still based on a wavelength comparison.

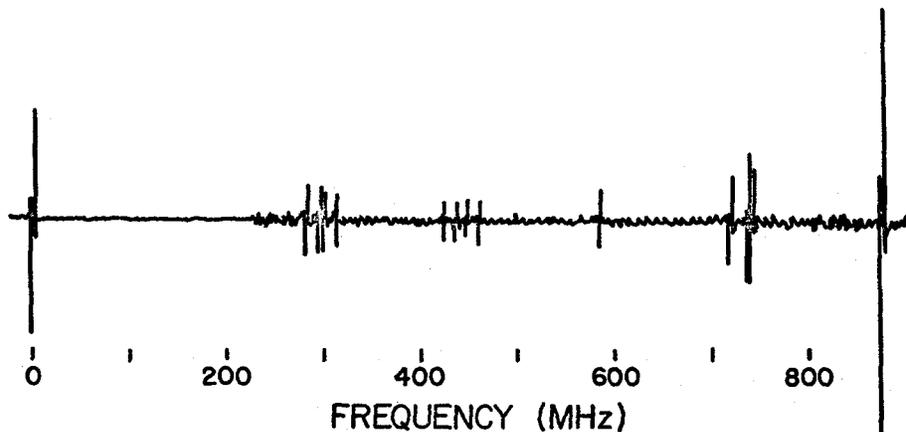


Fig. 9 I_2 hyperfine spectrum at 520 THz.

Even after successful operation of systems, such as that described, allows the accurate measurement of frequency standards in the visible, they will probably be limited for some time to widely spaced "bench marks"; special experiments will be necessary to perform the interpolation required to measure lines of particular importance, such as the H_α line at 656 nm used to measure the Rydberg constant. The convenience and accuracy, now associated with frequency measurement in the microwave region, will not be possible in the optical region without a considerable improvement in the tools available: tunable lasers, broad band nonlinear mixers and very high speed detectors. The best W-Ni point contact diodes have excellent properties of sensitivity and high non-linear coefficients and, if it weren't for their fragility and the uncertainties in the process of producing the best ones, the task of phase-locking up to about 200 THz would be almost straightforward. Attempts have been made to produce more uniform, rugged detectors by the use of vacuum deposition (11,12,13) but so far no practical device for the high frequency range has been demonstrated. A major problem has been to produce a sufficiently small contact area.

The author has recently made some experiments with vacuum deposited devices that showed very interesting preliminary results. The approach was based on two principles: (1) that only the area per unit length need be very small if irradiation on a line junction is coherent along its length; (2) that the narrow contact along the edge of a deposited film would provide small enough area per unit length (12). Devices were made by breaking glass substrates on which Ni had been deposited and subsequently evaporating Al as a counterelectrode on the broken edge, including the edge of Ni film which had been allowed to oxidize in air at room temperature; the Ni-NiO-Al contact area was estimated to be about 40 nm high by 200 μm long.

Preliminary results have shown that the diodes respond at room temperature to 30 THz radiation, giving typically S/N \approx 40 db for a 25 MHz beat signal between two laser outputs of about 100 mW each. A beat was observed between a Gunn oscillator output at \sim 40 GHz and the difference frequency, also about 40 GHz, produced by two CO₂ lasers operating on neighboring transitions. The devices also exhibited an interesting bistable switching phenomenon: when overloaded, whether by bias current or by laser radiation, the normal \sim 30 Ω resistance would switch to about 10⁸ Ω ; if subsequently a high voltage with limited current was applied, the device would revert more or less to its original state, both as regards its resistance and its response as a 30 THz detector. This work as well as some recent quantitative work on W-Ni point contact diodes by B. Whitford (submitted for publication to IEEE J. Quantum Electron.) suggests that at present the understanding of the mechanism of metal-oxide metal detectors for the THz region is in a very primitive state. Perhaps one can hope, without undue optimism, that great improvements in practical devices will follow with a better understanding.

Acknowledgements.- The author wishes to acknowledge that the work described in this paper is very much a collaborative effort by his colleagues G. Hanes, H. Riccius, K. Siemsen, D. Smith and B. Whitford, and depends on the capable technical assistance provided by W. Berger, L. Jones and R. Léonard.

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