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A Stabilized HCN Laser for Infrared Frequency Synthesis

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***Abstract*—Infrared frequencies have recently been synthesized in suitable diodes up to 88 THz with accuracies of parts in 10^9. Stabilized lasers are necessary in order to make frequency measurements of higher accuracy. The hydrogen-cyanide laser is the lowest frequency basis laser used in these synthesis schemes, and its stabilization has been the subject of recent interest. The laser is stabilized by locking it to a phase-locked microwave reference chain. Two servo loops are utilized. The first loop is a relatively slow frequency-lock loop with the correction applied to a piezoelectric-translator driver. This loop not only accommodates thermal expansion of the laser, but also serves as an acquisition aiding loop for the second servo. The latter is a phase-locked system with the correction applied to the laser discharge current controller. Data regarding the system stability are presented.**

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Introduction

An OBJECTIVE of infrared frequency synthesis [1] (IFS) at the National Bureau of Standards (NBS), Boulder, Colo., has been to determine the frequency of the 3.39-μm P(7) transition in methane. An example of infrared frequency synthesis is shown in Fig. 1. To accurately measure the frequency of a laser ν_M one must synthesize a frequency ν_s which is close to the frequency to be measured. The difference between these two frequencies is an intermediate frequency ν_{IF}, typically in the 10-100 MHz range. The synthesized frequency is

$$\begin{aligned}\nu_s &= \nu_M \pm \nu_{IF}\\ &= l\nu_1 + m\nu_2 + n\nu_3\end{aligned}$$

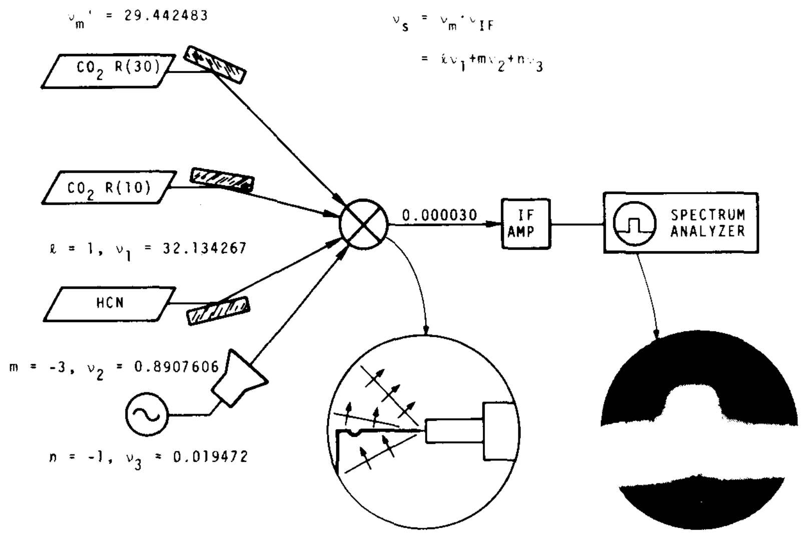


Fig. 1. Example of infrared frequency synthesis with HCN laser. All frequencies are given in terahertz.

where ν_1 and ν_2 are basis laser frequencies which have been determined by prior synthesis measurements, 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 which produces the intermediate frequency to be measured, occurs in a suitable diode—typically a tungsten catwhisker on a nickel base. Two relevant points are indicated in Fig. 1. First, the laser beams are polarized with the electric field vector in a plane which contains the catwhisker antenna while the angles between the laser beams and antenna are as dictated by antenna theory [2]. Second, the beat-frequency signals usually have not been of sufficient quality to allow cycle counting. One then determines the frequency of the beat-note center with respect to a frequency marker from a tracking generator by using a storage display for averaging the RF power spectrum of the beat. It is desirable to narrow the laser's power spectral linewidth to reduce the uncertainty in locating its center.

The hydrogen-cyanide (HCN) laser is the lowest frequency basis laser used in most IFS schemes, and its stabilization, along with the stabilization of the other basis lasers, has been a subject of recent interest. A group at Martin Marietta reported phase locking of an HCN laser for brief periods [3]. Another group at the National Physical Laboratory in Teddington, England, has frequency locked an HCN laser to a molecular absorption to a stability of parts in 10^8 [4]. Before we describe some HCN laser stabilization work at NBS, Boulder, we will first illustrate where it fits into the overall scheme of things.

The frequency of methane was recently measured at NBS to better than a part in 10^9. Fig. 2 shows the stabilized synthesis chain that was used in that series of measurements [5]. (The equations on the right relate the particular ν_M to the measurements preceding it.) The He-Ne laser was stabilized to the methane saturated absorption [6]. The two CO_2 lasers were stabilized to the R(30) and R(10) transitions [7]. The water-

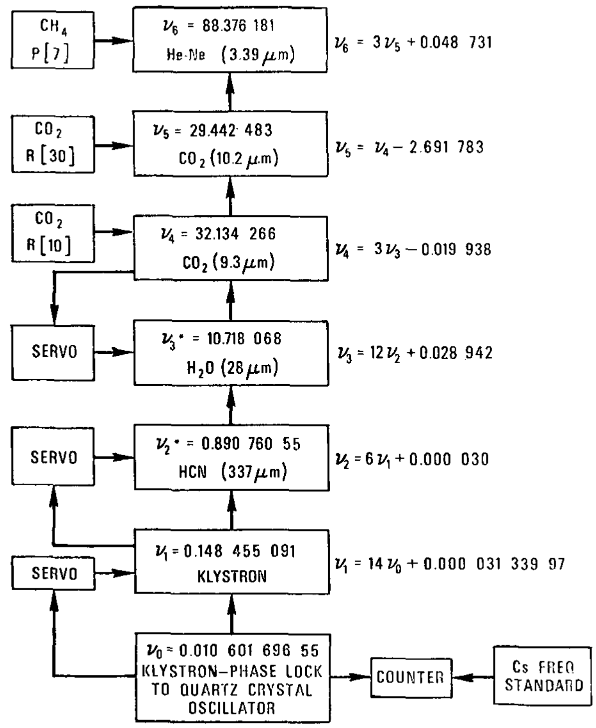


Fig. 2. Stabilized infrared frequency synthesis chain. All frequencies are given in terahertz. Wavelengths are stated in parentheses.

vapor laser was stabilized by frequency locking its third harmonic plus the microwave frequency to a frequency 60 MHz away from the R(10) line of a CO_2 laser.

The HCN laser was frequency stabilized by locking it to a phase-locked microwave reference chain whose frequency at X band was determined by a counter, which in turn was referenced to the frequency of a cesium primary standard via the AT (NBS) atomic time scale [8], [9], in the NBS Time and Frequency Division. We note that both the HCN and H_2O lasers serve as laser transfer oscillators in this scheme. Due to the big step between these two lasers (12 harmonics), the re-

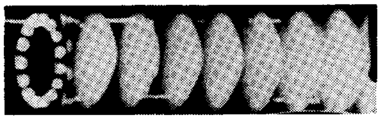


Fig. 3. New anode design and resulting stable plasma configuration in HCN laser. Striations are about 2.5 cm apart.

sulting beat note is weak and generally not sufficiently reliable to use in a servo loop. Hence, the frequency comparison between cesium and R(10) of CO_2 is made at this point, rather than transferring up or down in frequency.

Our next goal at NBS is to improve the methane frequency measurement to an accuracy of a part in 10^{10}. This future experiment is the motivation behind the HCN laser stabilization discussed here.

Passive Techniques

An accuracy of a part in 10^{10} requires knowledge of the HCN laser frequency to better than 90 Hz. Some of the problems encountered in this work and some of our solutions follow.

The laser is a folded confocal type 4 m in length and 10 cm in diameter. The usual passive stabilization techniques are used, such as Invar rods for spacers to reduce thermal drift and current-regulated power supplies for the laser discharge. The gas mixture is 0.150 torr (1 torr = 133.3 N/m^2) of methane and 0.275 torr of ammonia in a flowing system. This gives rise to a reasonably stable plasma at 0.6 A and produces stationary striations about 2.5 cm apart. This is desirable since an unstable plasma can cause frequency fluctuations through the resulting changes in the refractive index.

The power is coupled from the laser by a beam splitter. This is important for the HCN laser for several reasons. First, one can couple out a large power with minimum mode distortion [10]. In the scheme just shown in Fig. 2, sufficient power is required from the HCN laser to generate the 12th harmonic as compared with a maximum of 3 harmonics in all other cases. Second, the beam splitter polarizes the laser beam and provides better coupling to the diode than could a hole-coupled laser with its randomly changing polarization [2]. Third, one can replace the side mirror of the beam-splitter output laser by a polyethylene window and have an output from each side of the laser—the original one for stabilization and the additional one for synthesis or testing.

The beam-splitter coupling scheme does have its own peculiar problems, however [10]. In stabilizing the laser, the phase-lock correction is applied via current control. The current modulation causes an acoustic wave in the plasma and a vibrating membrane resonance in the 0.002-in (0.050-mm) beam splitter is excited when the frequency of the correction current is near 250 Hz. In order to eliminate this unwanted response, it was necessary to put in a 0.020-in (0.5-mm) thick polyethlyene beam splitter which dropped the power from 20 to about 12 mW [11].

Previous HCN laser designs had a simple stainless-steel ring for the anode and the plasma could fluctuate by terminating at different places on it. Fig. 3 shows the anode now in use, which electrically confines the plasma to certain regions near the anode. This aluminum electrode is anodized and is electrically insulated except on the 15 conducting inserts. These inserts are stainless-steel rods about 1 in long, which fit tightly into the holes for good electrical contact. This anode design leads to exceptional plasma stability.

Fig. 4. Qualitative tests of reference sources. Vertical scale is linear.

The laser, as well as the microwave reference components are all placed on a vibration-isolation table. All together, these passive techniques decrease the laser's linewidth from about 20 to about 2 kHz.

Reference Chain

An immediate objective is to obtain both long- and short-term reference stabilities of parts in 10^{10} at 1 THz. The basic reference chain is as follows [11]. An X-band klystron is phase locked to some harmonic (say the 100th) of a crystal reference; then an E-band klystron is locked to the 7th harmonic of the X-band source. The HCN laser frequency is then compared with the 12th harmonic of the E-band source. (An alternative scheme would go from 10.6 GHz to 0.891 THz using a Josephson junction [12]; however, the disadvantages outweigh the advantages at this time.)

The absolute stability of the locked laser depends upon the driving source of the reference chain, a quartz crystal oscillator. To synthesize from a good quality quartz oscillator at 5.06 MHz to the HCN laser at 0.891 THz requires a multiplication factor of 176 400. This places some rather stringent requirements on the spectral purity of the quartz oscillator's output [13].

Our most convenient qualitative check on the short-term stability of the reference oscillator (5–100 MHz) is obtained by examining the beat note between the multiplied reference signal and the HCN laser at 0.891 THz. When multiplied to this frequency, the short-term stability is sometimes such that the multiplied reference linewidth is broader than the laser linewidth (which was to be narrowed by locking to the reference). Fig. 4 shows an extreme example of this, as well as of some sources of noise. These linewidths are to be compared with the current 2-kHz linewidth of a state-of-the-art laser. The HCN laser is free running in these tests.

Fig. 4(a) denotes the beat note where the X-band oscillator is locked to a multiple of a synthesizer signal at 5.06 MHz. The dispersion is 1 MHz/division. The dispersion for the next three beat notes is 50 kHz/division. For Fig. 4(b), the reference is a

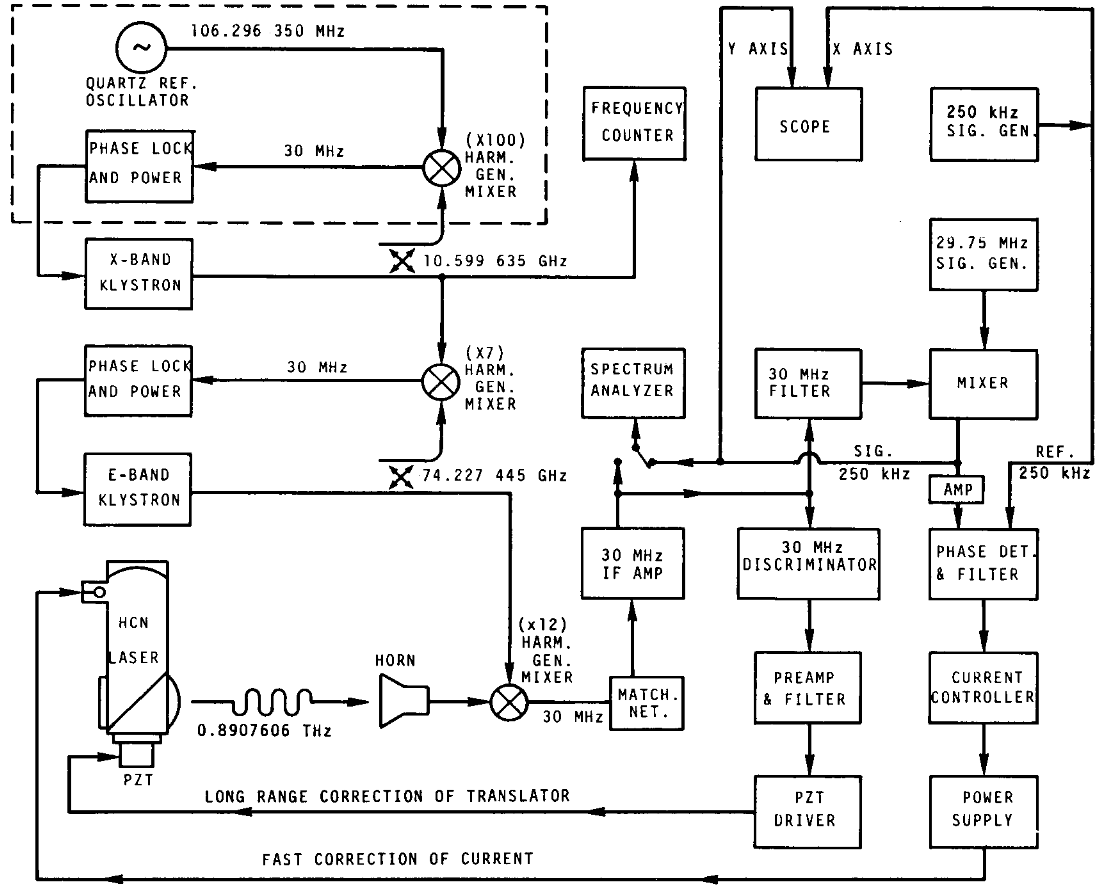


Fig. 5. Laser stabilization scheme.

5.06-MHz quartz oscillator. However, the multiplier chain going from 5.06 to 106 MHz was not state of the art. In Fig. 4(c), the beat note is referenced to a 106-MHz oscillator; and the last beat note in Fig. 4(d), results from a free-running Gunn diode oscillator at X band. (The last two beat notes are instrument limited by the spectrum analyzer due to the particular dispersion and resolution settings used here.) This sequence of beat notes suggests that a Gunn diode oscillator (when stabilized by a superconducting cavity [14]) may be the best reference for locking an HCN laser.

A question one might ask at this point is how much of the 2-kHz beat-note linewidth is due to the laser and how much is due to the multiplied reference? Some insight is provided by beating two free-running HCN lasers together. The resulting beat linewidth is about 4 kHz, which would indicate a width of not less than 2 kHz for each laser [13].

Experimental information regarding the reference width is unavailable at this time. Furthermore, the problem of obtaining a suitable reference has not been completely solved. A cavity-stabilized klystron (which will be stabilized in the long term via injection locking by a phase-locked klystron) hopefully will offer some improvement in this area. Calculations indicate a linewidth of less than 100 Hz could be achieved; however, this has yet to be tried. The results which follow in the remainder of this work were obtained by using a klystron which was phase locked to a quartz crystal oscillator at 118 MHz.

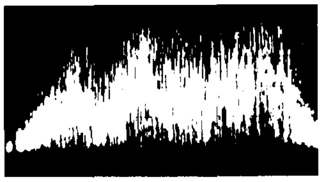
Active Techniques

Another of the purposes for beating the two lasers together was to obtain a signal with which to phase lock the two lasers at about 250 kHz apart. (At frequencies less than 200 kHz, one laser would injection lock the other.) One can then determine the filter parameters that lead to the best phase lock between the two lasers and then start with these values to lock the HCN laser to the microwave chain.

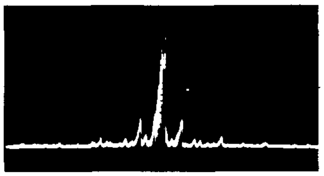
Fig. 5 shows the scheme for stabilizing the laser [11]. It consists of two servo loops: one for frequency locking and the other for phase locking. The frequency-lock loop consists of a frequency discriminator, preamp, and filter, and a translator driver with a slow (up to 100-Hz frequency response) correction applied to the piezoelectric translator (PZT). This loop serves as the acquisition aiding loop for the phase-lock system.

The phase-lock portion of the scheme is shown on the right. It consists of a filter, a 29.75-MHz quartz oscillator, a balanced mixer, a 250-kHz reference oscillator, a phase detector and filter, a current controller, and the power supply. The 30-MHz signal is mixed with 29.75 MHz, and the 250-kHz difference signal goes to the phase detector and to the Y axis of the scope. The X axis of the scope is driven by the phase-detector reference.

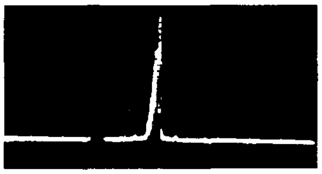
The phase-detector output is filtered and is fed as a correction signal to the pass tubes in the power supply which controls the current through the laser. The upper response fre-



(a)



(b)



(c)

Fig. 6. Beat notes between laser and microwave reference chain, down converted to 250 kHz. Vertical scale is linear, dispersion is 200 Hz/division, resolution is 20 Hz, and sweep rate is 5 s/div. (a) Laser is frequency locked. (b) Laser is phase locked. (c) For comparison with (b), the spectrum of 250-kHz reference to phase detector is shown. Dispersion, resolution, and sweep time are unchanged.

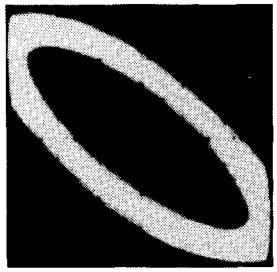


Fig. 7. 250-kHz Lissajous figure where Y axis is driven by down converted beat note and X axis is driven by phase-detector reference.

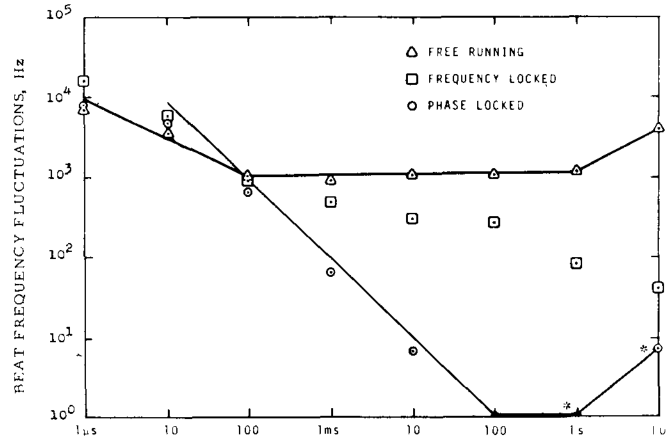


Fig. 8. HCN laser stability as measured with computing counter. Vertical scale is square root of Allan variance, $\sigma_{\delta\nu}{}^2(N, T, \tau, B)$ of the frequency fluctuations for $N = 2, T - \tau \approx 3$ ms, and $B = 200$ kHz.

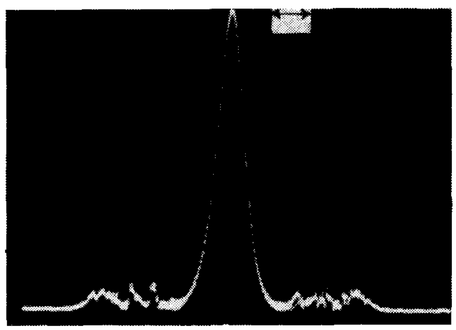


Fig. 9. Phase-locked beat note at 30 MHz. Vertical scale is linear, dispersion is 20 Hz/div, bandwidth is 10 Hz, and sweep rate is 2 s/div.

quency of the closed-loop controller/laser combination is about 5 kHz.

Fig. 6 shows the beat note between the HCN laser and the reference when the laser is frequency locked [Fig. 6(a)] and when the laser is phase locked [Fig. 6(b)]. Fig. 6(c) shows the phase-detector reference for comparison.

Fig. 7 shows a Lissajous figure where the Y axis is derived from the beat between the laser and microwave reference chain (down converted 250 kHz) while the X axis is driven by the phase-detector reference oscillator. We note here that there is an advantage to operating the phase-lock loop at 250 kHz, as, say, compared to 30 MHz, due to the fact that one can monitor the Lissajous figure while adjusting the filter parameters, gain, etc. for good operating values. The Lissajous figure also tells when the loop becomes unlocked and skips cycles. Of course, with a scope with a 30-MHz X-axis drive capability one could do the same.

Fig. 8 shows some preliminary indications of beat-frequency fluctuations (square root of the Allan variance [15]) as determined with a computing counter. These points represent 100 contiguous measurements, except for the two points indicated by an asterisk where $M = 10$. Shown are frequency fluctuations versus averaging time for the HCN laser under three different modes: free running, frequency locked, and phase locked. The rise at 10 s in the phase-locked mode is due to the laser dropping out of lock momentarily. The frequency-lock loop does cause it to return immediately to the phase-locked condition. We are working toward keeping it locked for longer periods, although it is not absolutely essential for our present purposes of IFS.

Fig. 9 shows the beat note at 30 MHz on a spectrum analyzer for the phase-locked condition. The post-detection bandwidth is greater than 10 kHz. The probability of seeing the beat note at the center of this 200-Hz window is better than 97 percent during a 10-min period, and thus the system could be utilized for IFS as is. If it is in the center of the window, we know its frequency to within about one part in 10^{11}. In general, one can count five or ten consecutive times for 10 s and determine the difference frequency when the beat note is as shown. (In the event that the signal is too weak to count, one can count the 29.75-MHz and the 250-kHz reference, the sum of which is the phase-locked beat frequency). From the data displayed we infer that the laser is following the reference but we still do not know how wide the laser line is.

We have an experiment planned to measure directly the linewidth of the reference (and consequently of the laser) at 1 THz. One would like to beat the 84th harmonic of one X-band klystron with the 84th harmonic from another tuned to a slightly different frequency, but there are ambiguities in such an experiment. However, by phase locking an HCN laser to the 84th harmonic of klystron A, all the other harmonics are discarded, and the 84th harmonic of klystron B can be compared with the phase-locked HCN laser with a Josephson junction multiplier mixer [12].

Discussion

The future direction of our work to improve the accuracy of the methane measurement will be decided after the linewidth W' of the phase-locked HCN laser has been determined. Calculations indicate that a linewidth of less than 100 Hz should result from the cavity-stabilized klystron on hand [11], and that a specially constructed multiplier chain with crystal filters at various stages could yield a width at 1 THz of 10 H, or possibly better [16].

One could then consider phase locking the water-vapor laser to the 12th harmonic of the HCN laser. This would lead to a 120-Hz-wide water-vapor laser linewidth, assuming that the laser noise is flicker of the frequency (the τ^0 dependence of the free-running laser in Fig. 8 indicates this), and hence the multiplied linewidth $\hat{W} = nW'$ [13]. A preferable approach would be to phase lock down from the CO_2 laser, where linewidths of 10 H have been reported for passively stabilized lasers [17]. This procedure, if successful, could lead to a water-vapor laser linewidth of 3 H. Furthermore, the phase error in the water-vapor laser signal is multiplied by a factor of three in the multiplication process and this would enhance the tightness of the phase lock. The same concept could be extended to the upper end of the chain. Conceivably, one could start at the methane stabilized laser and phase lock down to the water-vapor laser, phase lock the HCN laser to the cesium standard, and make the comparison at the $\times 12$ step. With both the HCN laser and the water-vapor laser phase locked, this beat note would be expected to be of sufficient quality for cycle counting and to yield a result of considerably improved accuracy.

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