Frequency Stability and Stabilization of a Chemical Laser

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Abstract—We have built a low-power CW HF/DF chemical laser, designed to achieve high-frequency stability. Measurements are reported which characterize the instantaneous spectral width of the laser output to less than one part in 10^{11} ($\Delta\nu < 1$ kHz) and the variations in absolute frequency of this emission with time to four parts in 10^{10} ($\Delta\nu = \pm 20$ kHz) per 0.1 ms. Two experiments to actively stabilize the laser frequency are reported. In one experiment the laser was locked to a high-finesse Fabry–Perot to five parts in 10^9 ($\Delta\nu = \pm 250$ kHz) for many minutes. In the other experiment one laser was locked to another using heterodyne beat spectroscopy to 1.7 parts in 10^9 ($\Delta\nu = \pm 85$ kHz). The stabilization experiments were limited by the feedback loops used.

I. INTRODUCTION

A frequency stabilized chemical laser is a very useful tool in many aspects of laser research and applications, including chemical laser diagnostics, spectroscopy, and laser radar. A laser radar using a chemical laser amplifier is especially attractive due to the excellent atmospheric transmission in the DF laser band. The present work was motivated by such an application, where a frequency stabilized laser is required to serve as a driver for the amplifier and as a local oscillator for the receiver. To be useful for some defense applications, the laser radar must have a reference laser with a minimum frequency stability of one part in 10^9 (total frequency excursion of 100 kHz) for periods of several milliseconds, or longer. The objective of the present work is to demonstrate that a CW chemical laser can be stabilized to $\Delta\nu/\nu = 10^{-9}$ for periods of time equal to or greater than 1 ms.

Prior to the present work, the chemical laser had been observed to operate on a single transition [1] but there were no measurements of the frequency stability characteristics of the laser. Concurrent with the initial stages of the present effort, several authors reported measurements of the frequency characteristics and active stabilization of chemical lasers. Eng and Spears [2] reported a relative stabilization of $\Delta\nu = \pm 500$ kHz (i.e., a total $\Delta\nu/\nu =$ one part in 10^8) on the $P_1(6)$ transition in HF by using a heterodyne lock to the second harmonic of a stabilized CO laser, and an absolute stability of ± 7 parts in 10^8.

Hinchen and Freiberg [3] reported a free-running frequency stability of ± 1 part in 10^8 per 150 ms by analyzing the laser output with a Fabry–Perot, and a stability of several parts in 10^8 by locking the laser to Lamb's dip. Wang [4] reported a free-running laser frequency deviation of 30 MHz per 100 ms, using the beat spectrum of two longitudinal modes in the laser.

In the present paper, we will report the results of our measurements of the frequency deviations and stabilization of a low-power CW chemical laser. These measurements show improvements of nearly an order of magnitude over the above results.

II. LASER DESIGN

The general problem of frequency stabilization of a laser has been summarized by Birnbaum [5]. The fractional variation in frequency of the output of a laser $\Delta\nu/\nu$ depends on the fractional change in the optical length of the laser resonator:

$$\frac{\Delta\nu}{\nu} = \frac{\Delta L}{L} + \frac{\Delta n}{n}$$

where L is the length of the resonator and n the index of refraction. Passive stabilization is accomplished by minimizing variations in L and n through careful engineering, the stability achievable by this method being limited mainly by vibration and thermal expansion of the resonator, and by density fluctuations in the medium. Active stabilization on the other hand is accomplished by detecting a change in the frequency, and subsequently adjusting L or n to remove that change. The stability achievable in this case depends on the minimum frequency change detectable, and the ability of the accompanying feedback loop to correct the change as quickly as it occurs.

In frequency stabilizing a laser one can follow one of two approaches. One method relies on improving the passive stability to the best practical level achievable, and then use a comparatively simple feedback loop to remove whatever minor instabilities and slow drifts remain. This method has been very successfully applied to the CO_2 laser [6]. The other method places the full burden of stabilization on the feedback loop, requiring a high gain-bandwidth in the loop to remove all frequency variations. This method has been very successful for dye lasers [7], [8]. Due to the uncertainties of the chemical laser frequency characteristics, we chose the first method for the initial careful laser design, and later applied more effort to the feedback loop.
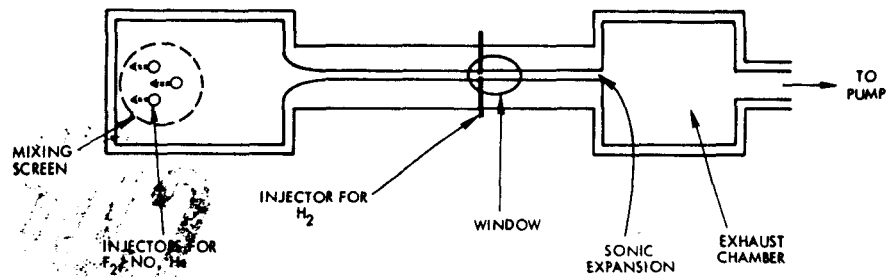
Fig. 1. Schematic of laser flow channel. Dimensions of the gain region
are 5 mm X 6 mm X 100 mm.

The laser concept selected was that of Shirley et al. [9] which is a purely chemical device. This concept was selected because control of density fluctuations within the lasing medium is a fluid mechanical problem only. In other designs [1] it was felt that the noisy electrical F_2 dissociation schemes would add unnecessary complications.

The laser flow channel was carefully designed to ensure a quiet, laminar, subsonic flow. It consists of three regions, the mixing chamber, the flow channel, and the exhaust region illustrated in Fig. 1. In the mixing chamber, fluorine and nitric oxide react in the presence of helium diluent to produce free fluorine atoms. The resulting mixture is accelerated down a laminar flow channel to the lasing region where hydrogen (or deuterium) is injected to react with the free fluorine, thereby producing the lasing inversion. Calcium fluoride windows, mounted at Brewster's angle, couple the gain medium to an external optical resonator. A nitrogen purge is provided to protect the windows from the fluorine. Typical operating conditions are: 3.9 mmol/s F_2, 0.1 mmol/s NO, 0.7 mmol/s H_2, 9.7 mmol/s He, and 1.0 mmol/s N_2. At a distance 37 mm downstream from the lasing zone, the temperature is 750 K and the pressure is 0.7 kPa (5 torr).

From the introductory remarks, it is clear that for good frequency stability, the index of refraction of the medium must be kept as constant as possible. The index of refraction of a gas depends upon the density, and it is easily shown that for a laser medium consisting primarily of He at 5 torr, a variation in density of 0.5 percent is sufficient to cause a laser frequency variation of one part in 10^9.

Variations in density will arise from turbulence in the flow, and this was independently investigated, using a hot wire anemometer. The instrument was capable of detecting density variations as small as 0.12 percent up to a frequency of 15 kHz. Density variations were found to be above the minimum detectable level at only two frequencies, namely 1.2 percent at 6 Hz, and 0.6 percent at 100 Hz, showing that turbulence had successfully been reduced to an acceptable level for the initial frequency stability goal.

The external optical cavity consists of two mirrors separated by 0.5-m long Invar rods. One mirror is a 4-m radius germanium substrate dielectric outcoupler mounted on a PZT crystal and the other is either a flat gold mirror for multiline operation, or a diffraction grating for single-line operation. Immediately in front of the grating is an adjustable aperture used for transverse mode control.

With the multiline configuration, simultaneous lasing on four to six transitions at a maximum outcoupled power of 1.4 W was observed. Single-line lasing on more than a dozen $P_1(J)$ and $P_2(J)$ lines with a maximum outcoupled power of 120 mW on $P_1(7)$ was observed using a Bausch and Lomb replica grating.

III. FREQUENCY CHARACTERISTICS

The laser was constrained to operate on a single frequency (single longitudinal and transverse mode) by using a 0.5-m long resonator and an intracavity aperture. By applying a sawtooth voltage to the PZT crystal on the laser, the output power as a function of frequency was observed, verifying single-mode operation, and displaying a prominent Lamb's dip (Fig. 2). Homodyne spectroscopy of the laser output, performed by spectral analysis of the output of a fast InAs detector illuminated by the laser showed a spectrum with detectable signal only below 400 Hz, and with a FWHM of less than 100 Hz. The spectrum agreed well with the slow, 2 percent amplitude variations in the laser output intensity, and confirmed that the laser was operating on only one independent optical frequency. This does not necessarily mean that the spectral content of the emission is a single frequency, since any nonindependent effects, such as, for example, a coherent frequency modulation, will not result in a beat in this experiment. However, in subsequent experiments, where two independent lasers were used to produce a heterodyne beat (see Section IV), it was shown that the spectral width of the emission of each laser is less than 1 kHz (one part in 10^{11}). The absolute frequency of this emission changes with time, and the stabilization effort amounts to confining these frequency excursions to within the bounds of the stabilization goal (100 kHz in this case). To construct an adequate stabilization system it is therefore necessary to measure the rate at which the frequency of the emission changes with time, or the Allan Variance [10] of the laser.

This measurement was carried out by using a high-finesse scanning Fabry-Perot interferometer built especially for the purpose. The interferometer consists of two mirrors, one flat and one concave (310-mm radius), separated by a 260-mm long quartz spacer. The mirror coatings were designed to have high reflectivity both at 2.8 μm and at 3.39 μm for absolute stabilization of the HF laser to a methane stabilized He–Ne laser (see Frequency Stabilization). The maximum experimentally measured finesse was 77 for the $P_1(4)$ transition (2.64 μm), and was found to vary from transition to transition
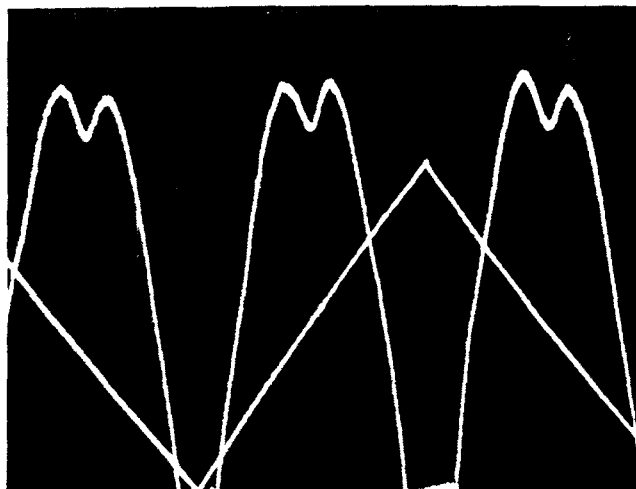
Fig. 2. Single mode and Lamb's dip in laser output. Full width of dip at half maximum is 20 MHz.

Fig. 3. Calibration curve for Fabry-Perot interferometer. Free spectral range is 500 MHz, finesse = 77.

due to variations in the reflectivity of the mirror coatings and absorption in the mirror substrates. With a finesse of 77, the FWHM intensity of the transmission peak can be calculated to be W = 7.5 MHz. From elementary considerations of the Fabry-Perot, the change in frequency of the laser $\Delta\nu$ can be related to the fractional change in transmitted intensity $(\delta I/I_{max})$ where I_{max} is the intensity transmitted at the peak of the fringe. If the laser line center frequency corresponds to the peak of the interferometer transmission, then

$$\frac{\delta I}{I_{max}} = 4\left(\frac{\Delta\nu}{W}\right)^2, \quad \delta I \ll I_{max}.$$

If instead the line is centered at the side of the transmission fringe, where the frequency discrimination of the interferometer is maximum, the frequency resolution is increased to

$$\frac{\delta I}{I_{max}} = \frac{\Delta\nu}{W}, \quad \delta I \ll I_{max}.$$

These results are plotted in Fig. 3 for the interferometer used. This figure is therefore the calibration of the amplitude to frequency conversion of the interferometer.

An experiment was performed to measure the frequency variations of the line center frequency as a function of the sampling time interval. The output of the laser was mode matched to the interferometer using a lens, and the laser was optically isolated from the interferometer using a linear polarizer and a quarter-wave plate. The laser was very slowly scanned through the transmission fringe of the interferometer by applying a ramp voltage to the PZT crystal on the laser. The intensity fluctuations in the transmitted intensity, occurring on the side of the transmission fringe were measured, and the corresponding magnitudes of the line center frequency fluctuations plotted against the time interval in which the variation was observed. For long time intervals (>0.1 s) the laser frequency varied an amount greater than the width of the transmission fringe, and the above method of gathering the data could not be used. Instead the laser was locked to the interferometer (see Frequency Stabilization) and the error signal required to keep the laser locked was calibrated in laser
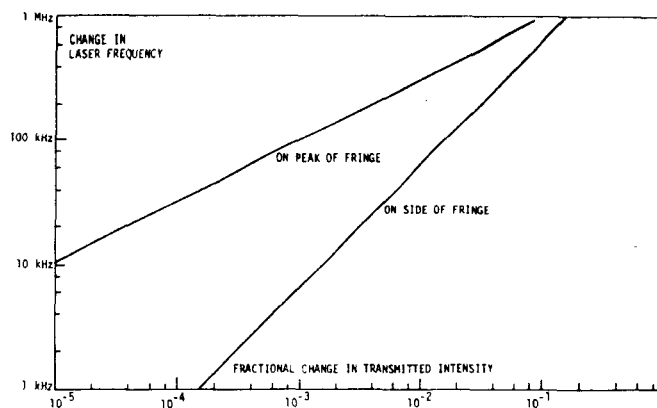
frequency deviations which would have occurred in the absence of a lock.

The results of these measurements are shown in Fig. 4. Also plotted on this figure are three points obtained in a later experiment when two separate HF lasers were used to produce a heterodyne beat signal, the frequency variations of which are proportional to the individual laser frequency deviations. This separate, direct frequency measurement, confirms the frequency calibration of the data obtained using the Fabry-Perot.

From Fig. 4, it can be seen that the laser has an average free-running frequency stability of about four parts in 10^{10} (40 kHz) per 100-μs interval. If the time interval is extended to one second, for example, the average frequency shift is about 40 MHz. The results shown characterize the frequency behavior of our chemical laser, and provide sufficient information to design an active stabilization system. For example, to obtain a stability of one part in 10^9, the feedback loop should have unity gain at a few kilohertz, and have sufficiently large gain at lower frequencies to cancel out the measured laser frequency variations.

IV. FREQUENCY STABILIZATION

To stabilize a laser, it is necessary to have a stable reference to which the laser can be locked. The stability achievable depends on the frequency discrimination characteristics of the reference source. Thus in the case of the 3.39-μm He-Ne laser, the stability of one part in 10^{12} can be achieved because the absorption peak in methane is very sharp: 150 kHz [11]. In the case of the HF laser, where any one of a number of different lines may be required, a reference which is less line specific is required. One possibility is Lamb's dip in the laser. We measured the width of Lamb's dip to be 20 MHz, which, combined with the amplitude fluctuations in the laser, limits the frequency stability achievable with this method to a few MHz. This is insufficient for the objective of the present effort. We therefore decided to use a high-finesse Fabry-Perot as a reference cavity.

There are several distinct methods by which a laser can be locked to an interferometer. The simplest method is an ac method, in which the laser is locked to the peak of the transmission fringe of the interferometer. If a sinusoidal dither voltage is applied to the PZT crystal on the interferometer, its
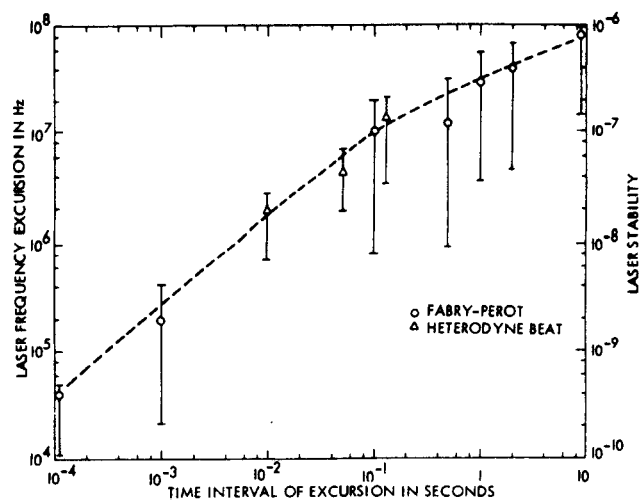
Fig. 4. Allan Variance of unstabilized laser. Magnitude of the laser frequency jitter is plotted against the time interval of the observation. Error bars are calculated for 95 percent confidence.

Fig. 5. Absolute stabilization of HF Laser. HF laser is locked to Fabry-Perot, which in turn is locked to a CH_4 stabilized He-Ne laser.

transmission frequency will change correspondingly. The difference in phase between the dither voltage and the resulting transmitted intensity can be detected using a lock-in amplifier and this signal can be used as an error signal and applied to the PZT crystal of the laser. In this manner, the laser can be locked to the Fabry-Perot. The transmission frequency of the Fabry-Perot will, however, change with time in response to environmental perturbations, such as, for example, changes in temperature. As a result, there will be a change in the absolute frequency of the lock, but this change may be removed by separately stabilizing the Fabry-Perot to an absolute frequency reference. This technique has previously been successfully applied to the absolute stabilization of dye lasers [13], and we shall briefly discuss the application of the method to the chemical laser.

The absolute frequency source used was a 3.39-μm He-Ne laser, stabilized to methane absorption cell. The laser is the design developed by Barger and Hall [11]. It consists of a He-Ne gain cell and a methane filled absorption cell inside the same laser resonator. By dithering the laser around the methane Lamb's peak an error signal can be generated to lock the He-Ne laser to the methane line center in the same way that was described above for locking the HF laser to the Fabry-Perot. With this method, an absolute stability of one part in 10^{12} has been achieved in the He-Ne laser [12].

To make use of this frequency stable source, the Fabry-Perot was designed to have a high finesse at both 3.39 μm and the HF band (2.6-2.9 μm). The measured values of the finesse were 60 at 3.39 μm and 77 at 2.64 μm. The Fabry-Perot was furthermore made from a large mass of quartz to reduce its sensitivity to perturbations, thereby facilitating its stabilization. To stabilize the Fabry-Perot the stabilized He-Ne laser emission was analyzed by the interferometer. In this case, any changes in transmitted intensity must be due to changes in the interferometer. If the Fabry-Perot is now dithered around the transmission peak of the He-Ne laser, and the error signal applied to the Fabry-Perot, the latter can be absolutely stabil-
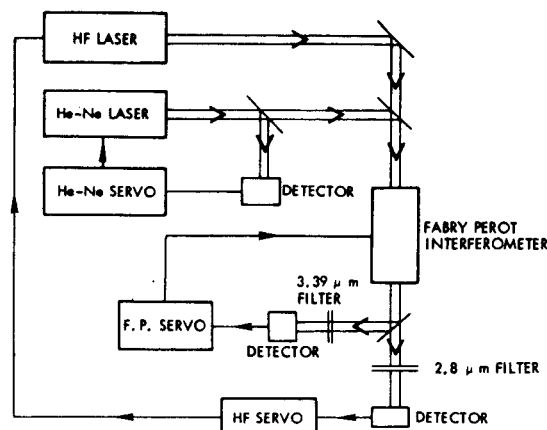
ized. By simultaneously locking the HF laser to this Fabry-Perot, as described above, the HF laser can be absolutely stabilized. The complete lock is illustrated in Fig. 5. For a given Fabry-Perot, the illustrated system will not permit the lock of the HF laser to an arbitrary absolute wavelength. This is because the interferometer must transmit both the He-Ne and the HF radiation, and the former is fixed. This restriction can be removed in practice by locking the Fabry-Perot to a simple He-Ne laser which in turn is locked to the methane stabilized He-Ne laser with an arbitrary frequency offset. (See below for details of such a lock when applied to two HF lasers.) In this manner the HF laser can be absolutely locked at an arbitrary frequency within the Doppler profile.

Our main goal in this work has been to demonstrate the applicability of these methods to the chemical laser, with emphasis on producing a suitable dual wavelength interferometer, and demonstrating the active stabilization of the HF laser. We have thus stabilized the Fabry-Perot to the methane stabilized He-Ne laser, and we have stabilized the HF laser to the same Fabry-Perot, thereby showing the feasibility of absolute stabilization of the chemical laser. We have not yet performed the simultaneous lock of both lasers to the interferometer. In what follows, we shall concentrate on the details of the lock of the HF laser, since it is the new and critical feedback loop. Due to the magnitude of the frequency variations of the chemical laser with time (Fig. 4) it is also the loop that is the most difficult to accomplish. The other feedback loops have previously been documented [12], [13].

The lock experiment was performed as described above, and a result from the lock is shown in Fig. 6. The HF laser was successfully locked to the Fabry-Perot for more than 20 minutes within frequency limits much smaller than the instrumental width of the interferometer. Small variations can be detected, but not accurately measured by this method. An upper limit of about 1 MHz can conservatively be claimed, but the exact quality of the lock must be separately investigated.

When the laser was locked to the Fabry-Perot, the latter was no longer available for measuring the frequency stability achieved. A calibration of the error signal can be used, but it is difficult to convincingly prove what part of the error signal
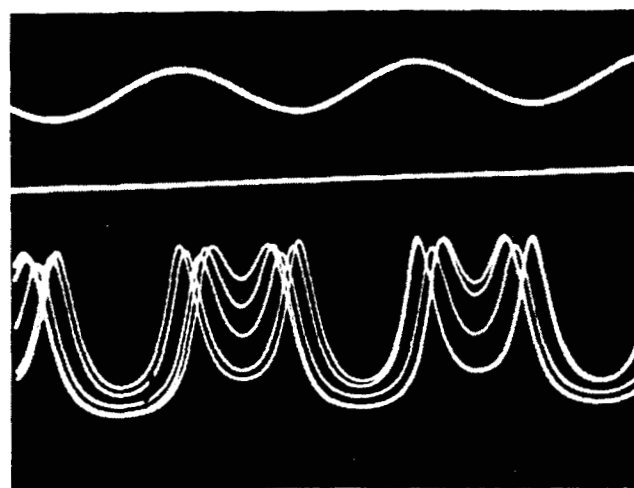
(a)
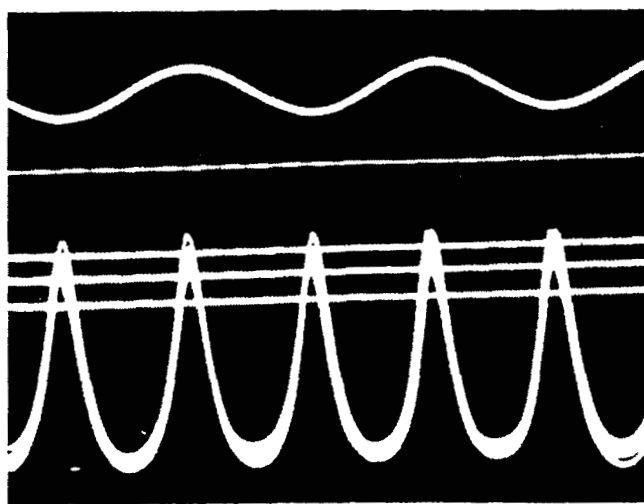
(b)

Fig. 6. Lock of HF laser to Fabry–Perot results. (a) Unlocked. Top trace: 500-Hz high-voltage dither on Fabry–Perot. Middle trace: high-voltage applied to laser. Bottom trace: transmission of Fabry–Perot on five consecutive sweeps, separated by 5 s each. FWHM of fringe is 7.5 MHz. Laser is drifting with respect to interferometer. (b) Locked. Top trace: 500-Hz high-voltage dither on Fabry–Perot. Middle trace: high-voltage correcting signal applied to laser. Bottom trace: transmission of Fabry–Perot four consecutive sweeps, separated by two minutes each.

is effectively used to correct the laser. The experimental verification of the frequency stability achieved was therefore carried out in a much more elaborate and convincing manner, by locking a second stable laser to a second Fabry–Perot and analyzing the variation in the difference frequency of the two locked lasers by heterodyne beat interferometry.

The experiment proved that each laser was locked to its interferometer to within a total excursion of 500 kHz (±250 kHz). Difficulties were encountered in improving the lock, due primarily to signal-to-noise limitations. From Fig. 3 it can be seen that to detect a change in frequency of 100 kHz, a change in transmitted intensity of one part in 10^3 must be detected. Due to losses in the Fabry–Perot, signal-to-noise problems did not permit achievement of this sensitivity thereby limiting the stabilization attainable. A further limitation was
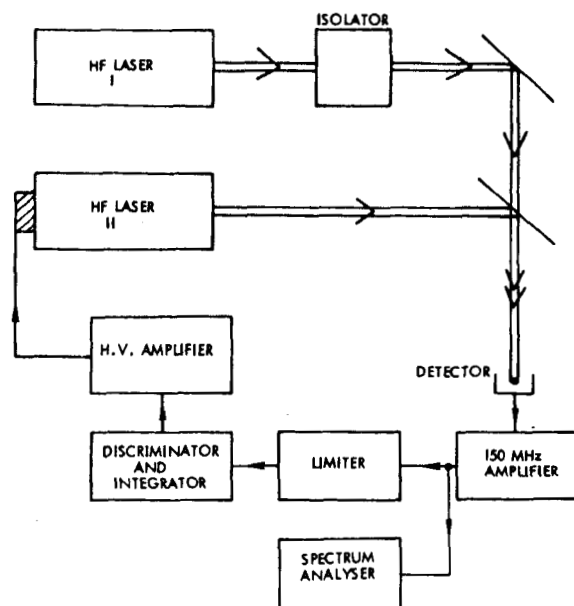

Fig. 7. Lock of one HF laser to another by heterodyne beat spectroscopy.

found to be in the bandwidth of the feedback loop. Although the dither frequency was up to 3 kHz, the bandwidth of the high-voltage amplifier and the PZT crystal limited the effective bandwidth to approximately 300 Hz.

The sensitivity of the lock system can evidently be increased by changing the electronics to a dc loop, locking the laser to the side of the fringe, where a change in transmitted intensity of only one part in 65 must be detected for a stability of 100 kHz. However, due to major changes in electronics required, this lock was not attempted. Instead another type of lock was performed in which one HF laser was locked to another by using the frequency of the heterodyne beat between them to produce an error signal. The experimental arrangement is illustrated in Fig. 7. The two independent lasers simultaneously illuminate a fast InAs detector (Judson J-12LD), the bandwidth of which is 150 MHz. A frequency discriminator, consisting of a double balanced mixer and two different lengths of coax cable, converts the change in beat frequency to a voltage which is amplified and applied to the PZT crystal of one laser to lock the two lasers together. Simultaneously, the beat frequency is analyzed using a spectrum analyser (Hewlett-Packard 851A/8551A). A typical result of this experiment is shown in Fig. 8. The total width of the spectrum is 250 kHz/30 ms. The frequency deviation of one laser with respect to the other is therefore $250/\sqrt{2} = 170$ kHz, or a deviation around a mean frequency of ±85 kHz. The offset frequency between the two lasers was 54 MHz, determined by the discriminator adjustment.

V. CONCLUSION

From the above experiments, we conclude that the chemical laser output has an instantaneous spectral width of less than 1 kHz, the absolute frequency of which varies with time as the resonance frequency of the laser resonator changes. These
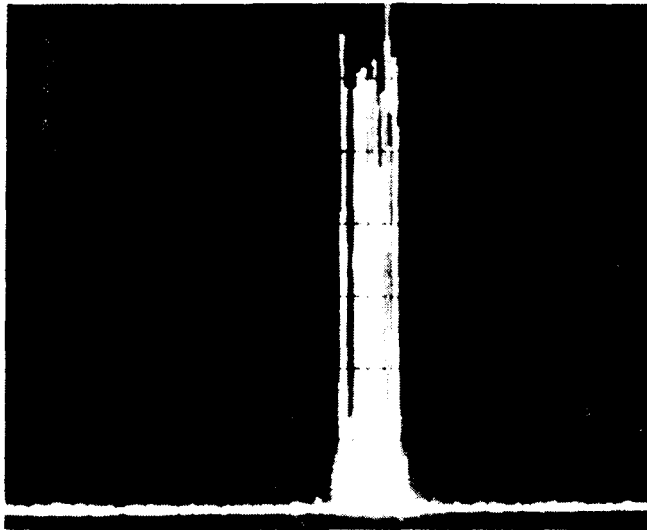
Fig. 8. Results of heterodyne beat lock. Horizontal scale: 300 kHz/cm and 30 ms/cm. If frequency: 10 kHz. Offset frequency between two lasers: 54 MHz.

variations display a well-behaved Allan Variance, thus permitting effective frequency stabilizating using a feedback loop. Two active stabilization experiments were performed. One demonstrated the means by which the HF laser can be locked to a Fabry–Perot interferometer, the stability achieved being ±250 kHz/many minutes, and limited by signal noise and by the bandwidth of the feedback loop. In another experiment, one chemical laser was locked to another using the heterodyne beat between the two lasers as the error signal. This feedback system was more sensitive than the first, and permitted a locked stability of ±85 kHz/30 ms. In both cases, the stability achieved was limited by the feedback loops rather than the chemical laser.

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