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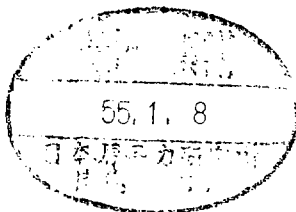
A Twin Optically-Pumped Far-Infrared
CH₃OH Laser for Plasma Diagnostics

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Abstract

A twin optically-pumped far-infrared CH₃OH laser has been constructed for use in plasma diagnostics. The anti-symmetric doublet due to the Raman-type resonant two-photon transition is reproducibly observed at 118.8 μm. With the 118.8-μm line, it is obtained from the frequency separation of the anti-symmetric doublet that the CH₃OH absorption line center is 16 ± 1 MHz higher than the pump 9.7-μm P(36) CO₂ laser line center. It is shown that the Raman-type resonant two-photon transition is useful in order to get several-MHz phase modulation for the far-infrared laser interferometer. Some preliminary performances of this twin laser for the modulated interferometer are described.

I. Introduction

In the modulated far-infrared (FIR) laser interferometer used for measuring the radial and temporal behavior of electron density of large Tokamaks [1], there are two different phase modulation techniques such as the mechanical Doppler shifting scheme by Véron [1, 2] and the non-mechanical dual beam scheme by Wolfe et al. [3]. With the mechanical modulation, although the maximum Doppler shifted frequency that has been achieved by Peterson et al. [4] is about one MHz, low frequencies of a few tens kHz are being used conveniently [1, 2, 5]. In order to realize several-MHz modulation stably, it may be important to know the cavity detuning characteristics of the optically pumped FIR laser [6] in detail. For such purpose, the anti-symmetric doublet due to the Raman-type resonant two-photon transition, which has been observed by Seligson et al. [7], and Heppner et al. [8, 9], may be attractive because it can provide us a wide frequency range of lasing.

We have constructed a twin optically-pumped FIR CH_3OH laser composed of two identical waveguide resonators for the modulated interferometer system, which will be applied, for example, to the JIPP T-II Tokamak machine [10], and the Raman-type resonant two-photon transition at $118.8 \mu\text{m}$ as well as at $70.5 \mu\text{m}$ was experimentally investigated in most detail. The pump offset frequency with the $118.8\text{-}\mu\text{m}$ line was obtained for the first time through the two-photon transition. Heterodyne beat signals were also observed to know the frequency stability of this twin laser.

Recently, Plainchamp [11] has investigated the frequency

instability of the two optically-pumped FIR CH₃OH waveguide lasers.

II. Experimental

Figure 1 shows the twin CO₂-laser-pumped FIR CH₃OH laser and the modulated interferometer attached to it for simulation experiments. The twin laser is of waveguide type [12], consisting of two identical 30-mm ID fused-quartz tubes of 2 m in length with plane external reflectors at each end, similar to the one by Hodges et al. [13]. Since the theoretical attenuation coefficient for the fused-quartz and for the linearly-polarized EH₁₁ mode is given by [12, 14]

$$\alpha = 1.827 \lambda_0^2 / r^3 \quad \text{dB/m}, \quad (1)$$

where λ_0 is the wavelength in m and r is the radius of cylindrical waveguide in m, then the calculated loss for the 30-mm ID fused-quartz tube of 2-m long is 0.35%/pass at 118.8 μm . This twin laser is set on a granite bench, and the separation between the laser mirrors is fixed using "Neoceram" glass pipes [15] having thermal expansion coefficient of maximum $\pm 1 \times 10^{-7}/^\circ\text{C}$. Although the input and output flat-mirrors, having a 2-mm hole at the center, were used in the heterodyne-beat experiments as shown in Fig.1, the 4-mm hole input mirror to introduce all the pump power into the cavity and the capacitive mesh output coupler [3, 16] to get as large diameter output beam as 30 mm in diameter were also tried. Each laser mirror is set between two bellows in order to

be free from atmospheric pressure changes. The output coupler assembly is located on a translation stage driven by a differential micrometer screw. On taking the detuning characteristics of the anti-symmetric doublet, the output coupler assembly was moved back and forth along the laser axis at the constant speed of 2 $\mu\text{m}/\text{min}$. The frequency change, $\Delta\nu$, on the detuning curve was calculated by simply using the tuning equation

$$\Delta\nu = -\nu \frac{\Delta L}{L}, \quad (2)$$

where ν is the lasing frequency, L is the cavity length, and ΔL is the fractional change of L . The pump CO_2 laser beam was injected into the waveguide through the coupling hole after being collimated [13] using an inverted telescope consisting of two ZnSe lenses with 1/4 magnification. The CH_3OH vapour flowed through the waveguide, and its pressure was measured by a Pirani gauge; the true pressure was about 40% of its indicated numbers.

The pump cw CO_2 laser with the free-spectral range of 80 MHz, built in our laboratory, can lase on the 9.7- μm P(34) and P(36) lines with maximum output power of 25 W and 23 W, respectively. Since variable frequency locking of the pump CO_2 laser was essential to observe clearly the Raman-type resonant two-photon transition, the pump CO_2 laser frequency was stabilized servo-mechanically (Fig.2) or manually by utilizing the slope of its detuning curve, as shown in Fig.3. This stabilization mechanism of non-modulation type is simple, easy to construct and inexpensive (a similar mechanism [17] has been reported independently after

this our system had been completed). The red He-Ne laser beam was combined colinearly with the invisible CO₂ laser beam at the ZnSe beam splitter for conveniently aligning the optical elements.

This twin CH₃OH laser can lase at the wavelengths of 70.5 to 699.4 μm. To get the tuning curves, a Golay detector or a pyroelectric detector were used.

The simple Mach-Zehnder type interferometers were composed, using 16-μm Mylar beam splitters for the simulation experiments. The heterodyne beam signals were generated in a Ge:Ga detector with two elements and displayed on a spectrum analyzer (HP 8553B/8552B). The Ge:Ga detector [18] had a maximum responsibility of over 10³ V/W and we could detect the heterodyne beat signals up to 6 and 5 MHz at 118.8 and 170.6 μm, respectively.

III. Experimental Results and Discussion

When there was some pump offset, the anti-symmetric doublet due to the Raman-type resonant two-photon transition was reproducibly observed at 118.8 μm, as shown in Fig.4, where the plus and minus frequency scans were taken to check the reproducibility while the pump CO₂ frequency was maintained constant (see Fig.3). Since the Doppler width at 118.8 μm is calculated to be 5.5 MHz (FWHM) at 300°K and the pump power coupled into the cavity was estimated to be on the order of one watt, the small width of about 2.3 MHz, which is mean value of two opposite frequency scans for the resonance pumping (in this case, CH₃OH pressure was 80 mtorr) may be pressure-broadened. This means that we are making the Doppler-free stimulated-emission

spectroscopy which will be discussed later. With the anti-symmetric doublet, the large (small) peak corresponds to forward (backward) gain [7]. The doublet, which appears on the FIR laser transition when there is a pump offset, comes from the Doppler effect; and the directional anisotropy in the FIR gain is due to the quantum mechanical feature of the optically-pumped three-level laser where both pump- and FIR-transitions are Doppler-broadened [7]. The doublet separation frequency, δ , is given by

$$\delta = 2 \cdot |\Delta| \cdot \frac{\lambda_{\text{pump}}}{\lambda_{\text{FIR}}}, \quad (3)$$

where Δ is the pump offset frequency which is defined as pump CO_2 laser frequency minus CH_3OH absorption line center frequency, and λ_{pump} and λ_{FIR} are the pump-laser and the FIR-laser wavelength, respectively [7]. When the sign of pump offset is plus (minus), the larger peak corresponding to the forward gain is on the plus (minus) frequency side from the smaller peak, which clearly agrees with the theoretical prediction [7]. On the 118.8- and 170.6- μm lines, the CH_3OH absorption line center frequency was measured to be 15 ± 5 MHz higher than the CO_2 laser line center by changing the CO_2 laser frequency while the 170.6- μm laser line was set at the output peak, because the 118.8- and 170.6- μm lines share the same upper level as assigned by Henningsen [19] and Danielewicz and Coleman [20] and the 170.6- μm line does not have the doublet (Fig.6). Therefore, for on resonance pumping, the pump frequency was set around + 15 MHz from the CO_2 line center. With the right two tuning-curves on

Fig.4, the pump offset calculated to be - 11 MHz from the observed doublet separation of 1.8 MHz agreed within experimental errors with the actual pump offset of $- 14 \pm 5$ MHz; and with the left two ones, the pump offset calculated to be + 10 MHz (= + X MHz) using the doublet separation observed agreed also with the actual pump offset. For the anti-symmetric doublet, the ratio of the forward peak to the backward peak with the plus pump offset is larger than the one with minus pump offset. This may be understood as follows. In the case of plus pump offset the pump power is small because the CO₂ laser is set on the lower slope; so that the backward gain becomes not much larger than the cavity losses, resulting in smaller output power as pointed out by Seligson et al. [7].

As expected from eq.(3), the doublet separation at 118.8 μm increased continuously with increasing pump offset frequency, as shown in Fig.5. This means that the peak frequencies of FIR laser are tunable within a limited frequency range which largely depends upon the pump intensity as seen in Fig.9; that is, if the pump intensity is large, this tunable frequency range becomes wide because of high gain. Figure 6 shows that the anti-symmetric doublet is not present at 170.6 μm , where the pump offset from the absorption line center was maintained to be - 14 MHz and the both frequency scans were taken to show again the reproducibility. This may be due to the smaller δ at 170.6 μm being masked by collisional broadening [7].

Figure 7 shows the doublet due to the Raman-type resonant two-photon transition observed on both modes a and b at 70.5 μm ,

and a doublet separation up to 4.3 MHz was observed by changing the pump offset frequency. In this case, the pump intensity was such that six modes were allowed to lase at $70.5 \mu\text{m}$ and a single mode at $699.4 \mu\text{m}$. With the $699.4 \mu\text{m}$ line, no doublet was observed on the detuning curve. In Fig.7, almost symmetric doublet at the pump offset of -15.5 MHz which was calculated using eq.(3), where the pump CO_2 laser was nearly at the line center, is not clearly understood and left for further experimental and theoretical studies.

When the pump CO_2 laser was set at its line center manually, because the stabilization technique shown in Fig.2 can not work in principle on the top of the detuning curve, the anti-symmetric doublet with 2.6-MHz separation was observed at $118.8 \mu\text{m}$, as shown in Fig.8. In order to check that this doublet truly originates not from two adjacent waveguide modes but through the two-photon transition, the self beat signal was searched for in vain, using the Ge:Ga detector at all frequencies within the lasing region of 6.4 MHz. From the doublet separation and the fact that the larger peak is on the lower frequency side, the pump offset was calculated to be $-16 \pm 1 \text{ MHz}$, which is a good agreement with the value of $-15 \pm 5 \text{ MHz}$ as described above. However, it may be worth to note that the detuning curve of the cw CO_2 laser becomes greatly asymmetric with decreasing the diameter of laser tube to 8 mm as in this work, and it was impossible to determine exactly the line center frequency of the CO_2 laser. On the other hand, Leite et al. [21] have measured the pump offset to be $-25 \pm 5 \text{ MHz}$ by means of Lamb-dip

spectroscopy and Inguscio et al. [22] have obtained $- 29 \pm 3$ MHz by means of the IR-FIR transferred Lamb-dip spectroscopy (Lund et al. [17] have independently observed the transferred Lamb-dip). Independently, Lourtioz et al. [23] have measured it to be about $- 25$ MHz through the Raman-type resonant two-photon transition. Those different values for the pump offset with the $118.8\text{-}\mu\text{m}$ line may be mainly due to the uncertainties as finding the CO_2 line center frequency. Therefore Fig.8 may suggest that more accurate pump offsets for shorter lasing wavelengths than about $100\ \mu\text{m}$ can be obtained by using a pump CO_2 laser whose frequencies are locked exactly on the line center utilizing the Lamb-dip type resonances at $4.3\ \mu\text{m}$ [24]. Recently, Plainchamp [11] has measured the pump offset with the $118.8\text{-}\mu\text{m}$ line to be $- 16$ MHz utilizing the top of the saturated fluorescence Lamb-dip as a standard frequency, which is a good agreement with our value of $- 16 \pm 1$ MHz. In this way, if the absorption line centers of lasing molecules are measured, this will become one of the novel techniques in FIR molecular spectroscopy. Similar Doppler-free stimulated-emission spectroscopy has been done for the first time by Koffend et al. [25] with the optically-pumped infrared I_2 laser.

When the pump intensity was about one order of magnitude larger than in Fig.4, using the 4-mm hole input mirror to introduce all the pump power into the cavity, the power broadened anti-symmetric doublet was observed at $118.8\ \mu\text{m}$ (Fig.9) and a maximum doublet separation of 5.3 MHz was achieved. The 4.8-MHz width at $\Delta \approx 0$ MHz is power-broadened by 1.4 MHz since the

collision-broadened width is estimated to be 3.4 MHz where the CH_3OH pressure was 0.12 torr and the coefficient of pressure-broadened width of 28 MHz (FWHM)/torr by Heppner and Weiss [26] is used. Figure 10 shows how the power broadening contributes to shape of the anti-symmetric doublet at $118.8 \mu\text{m}$, where the CH_3OH pressure of 120 mtorr and the pump offset of - 16 MHz were maintained constant. By decreasing the pump power slightly in order to neglect the effects of cavity loss on FIR gain as seen in Fig.4, the width of both forward and backward peaks became slim, resulting in enhancement of the degree of asymmetry of the doublet. Figure 9 is plotted on Fig.11, where the pump offsets calculated from the measured doublet separations are adopted. In order to get wider frequency range of lasing and larger output power for the modulated FIR laser interferometer, a pump offset around - 7 MHz may be suitable. This shows the variable frequency locking with no frequency modulation of the pump CO_2 laser using, for example, a Stark cell [27] outside the CO_2 laser cavity, may be suitable; it is because as stated previously the detuning curve of the cw CO_2 laser utilizing the 8-mm ID tube becomes strongly asymmetric against linear mirror displacement, as seen in Fig.2 in ref.[28].

When a capacitive mesh coupler (grid constant and square separation being $76 \mu\text{m}$ and $21 \mu\text{m}$, respectively [3, 16]) was used for 10-W pump power, the width of 3.9 MHz (FWHM) was obtained on resonance pumping, and a few mW output power was observed. It is expected that this output power will be easily increased [13] by using the more powerful cw CO_2 laser which is now under

construction for the plasma diagnostics. Also, to get large cw output power near 120 μm it will be attractive for us to use efficient lasing molecule such as CH_2F_2 which has been found by Danielewicz et al. [29].

Figure 12 shows the about one MHz heterodyne beat signal with the free-running twin laser and with the weak pumping of a few watts. The single beat signal on Fig.12 clearly shows that the twin laser is lasing at a single transverse and longitudinal mode. In Fig.13, the long term fluctuation of the beat frequencies for this twin laser is shown, indicating that the beat signals are within ± 0.1 MHz for about 2 seconds. This may come from both the mechanical vibrations and the pump CO_2 laser power fluctuations of $\pm 10\%$ at 120 Hz. This will be improved in the near future by using more rigid structures and the stable pump CO_2 laser. Figure 14 shows one example of the long term drift of the beat frequencies for 24 minutes when the room temperature was $20 \pm 1^\circ\text{C}$, the pump beam being chopped at 20 Hz with 50% duty cycle. This shows that the about one-MHz beat frequency is stable to within ± 0.1 MHz, which is the same as in Fig.13. It turned out that from observing the long term drift to the heterodyne beat signals such as Fig.14, both "A" and "B" FIR lasers composing the twin laser (see Fig.1) expanded or contracted with a same rate under the room temperature change of $\pm 1^\circ\text{C}$; thus a beat frequency setting was almost constantly maintained for about half an hour.

IV. Conclusion

Our experiments have shown that the Raman-type resonant two-photon transition may be useful in order to get several MHz phase modulation for the FIR interferometer. When the pump 9.7- μm P(36) CO_2 laser is set at its line center, the pump offset frequency with the 118.8- μm line has been obtained for the first time to be -16 ± 1 MHz from the frequency separation of the anti-symmetric doublet. This twin FIR laser will be applied to, for example, the JIPP T-II Tokamak machine [10] to measure the temporal behavior of electron density in the near future, as will be published elsewhere.

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Figure Captions

- Fig.1 Schematic of modulated FIR laser interferometer system using the twin optically-pumped 118.8- μm CH_3OH laser.
- Fig.2 Experimental arrangement for the simple frequency stabilization with no frequency modulation of the cw CO_2 laser for the FIR laser pumping.
- Fig.3 The cw CO_2 laser frequency of P(36) line at 9.7 μm was stabilized servo-mechanically by utilizing the slope of the detuning curve. By changing the reference voltage in Fig.2, the lasing frequency was easily changed.
- Fig.4 Anti-symmetric doublet due to the Raman-type resonant two-photon transition observed at 118.8 μm of CH_3OH laser, where $X = 10$ which is the calculated value. From left to right, the CH_3OH pressure was 115, 80 and 91 mtorr, respectively.
- Fig.5 Doublet separation frequency at 118.8 μm was continuously increased with increasing the pump offset frequency. From right to left, the pump offset calculated is + 6.7, + 8.0 and + 9.8 MHz, respectively. The CH_3OH pressure was 120 mtorr.
- Fig.6 Anti-symmetric doublet was not present at 170.6 μm . The CH_3OH pressure was 91 and 95 mtorr at 118.8 and 170.6 μm , respectively.
- Fig.7 Doublet due to the Raman-type resonant two-photon transition observed on both modes a and b at 70.5 μm , where the CH_3OH pressure was 74 mtorr.

- Fig.8 Anti-symmetric doublet observed at $118.8 \mu\text{m}$ when the pump CO_2 laser was set at line center. The pump CO_2 laser power was a few watts and the CH_3OH pressure was 120 mtorr. The pump offset was estimated to be -16 ± 1 MHz from the doublet separation.
- Fig.9 Anti-symmetric doublet observed at $118.8 \mu\text{m}$ when the pump power was around 10 W. The pump offsets are calculated from the doublet separations. The doublet separation of 2.6 MHz corresponded nearly to the peak of CO_2 laser output power. The CH_3OH pressure was 120 mtorr.
- Fig.10 By changing the pump power intensity slightly in order to neglect the effects of cavity loss on FIR gain, the power-broadening effect was examined, where the CH_3OH pressure of 120 mtorr and the pump offset of -16 MHz were maintained constant.
- Fig.11 Frequency and output power vs. pump offset, observed at $118.8 \mu\text{m}$ when the pump intensity was large (see Fig.9).
- Fig.12 Heterodyne beat signal at 0.94 MHz with the free-running twin $118.8\text{-}\mu\text{m}$ CH_3OH laser.
- Fig.13 Long term fluctuation of the beat frequencies for the free-running twin $118.8\text{-}\mu\text{m}$ CH_3OH laser.
- Fig.14 Long term drift of the beat frequencies at $118.8 \mu\text{m}$ when the room temperature was $20 \pm 1^\circ\text{C}$. The pump CO_2 laser beam was chopped at 20 Hz with 50% duty cycle.

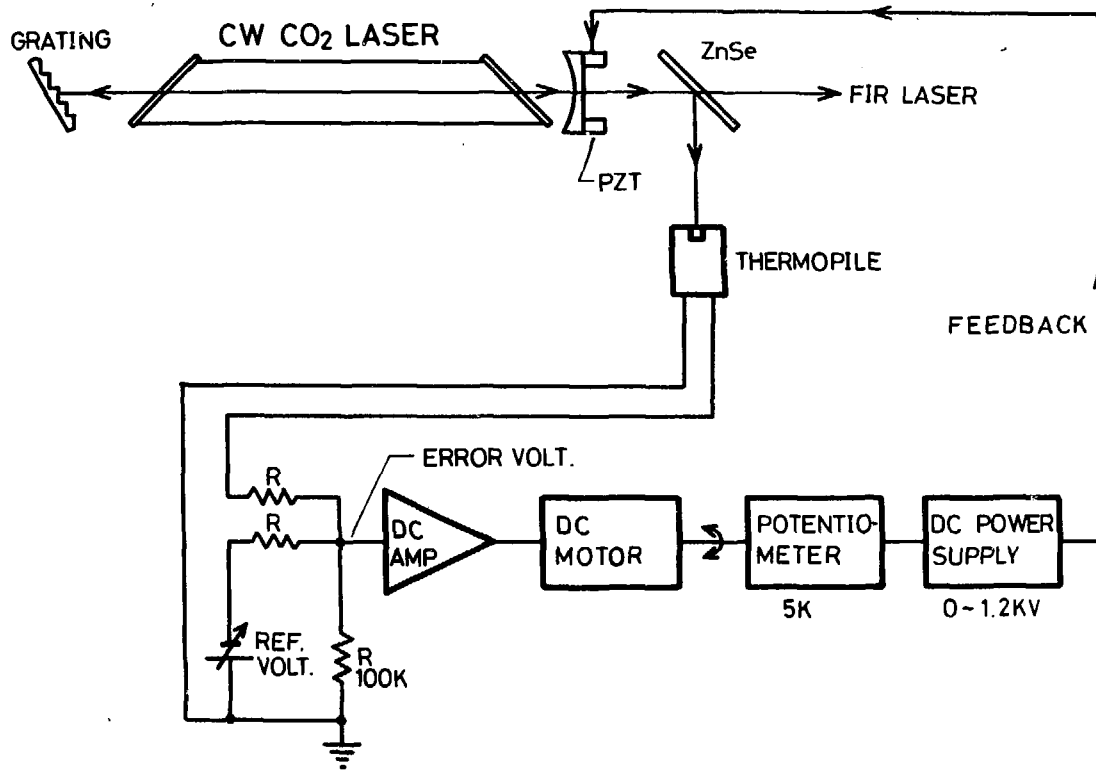


Fig.2

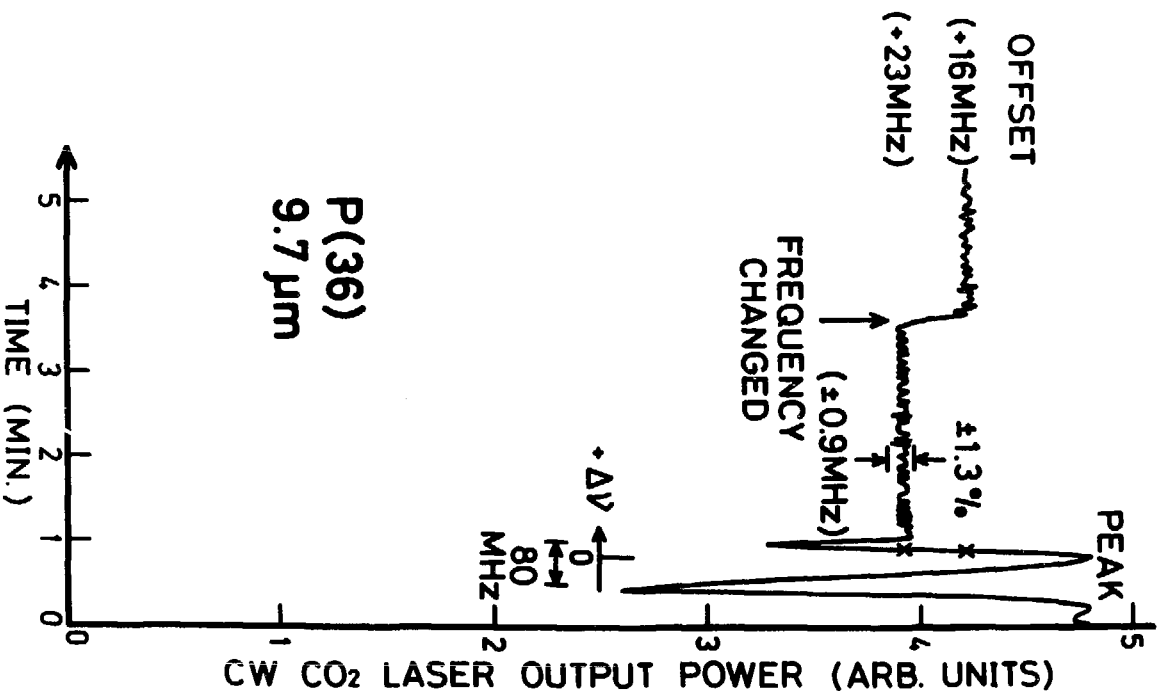


Fig. 3

118.8- μm CH₃OH LASER

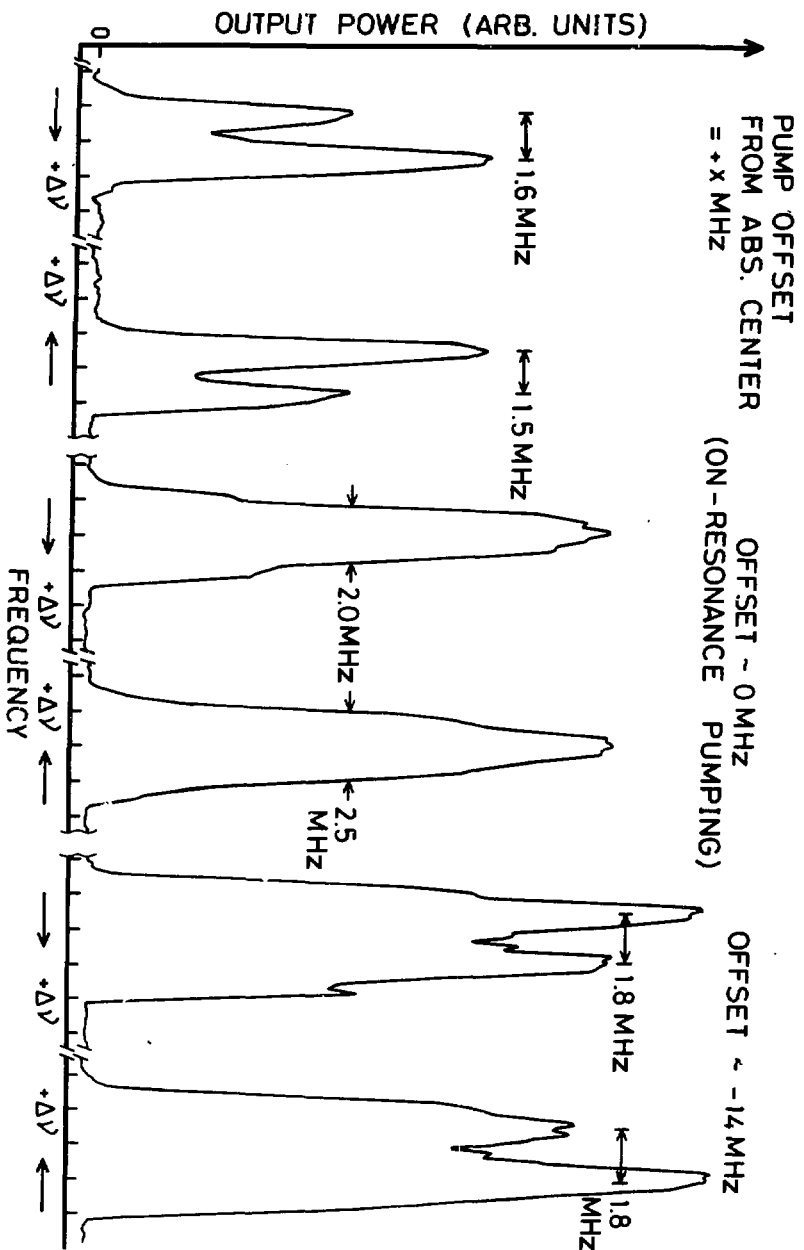


Fig. 4

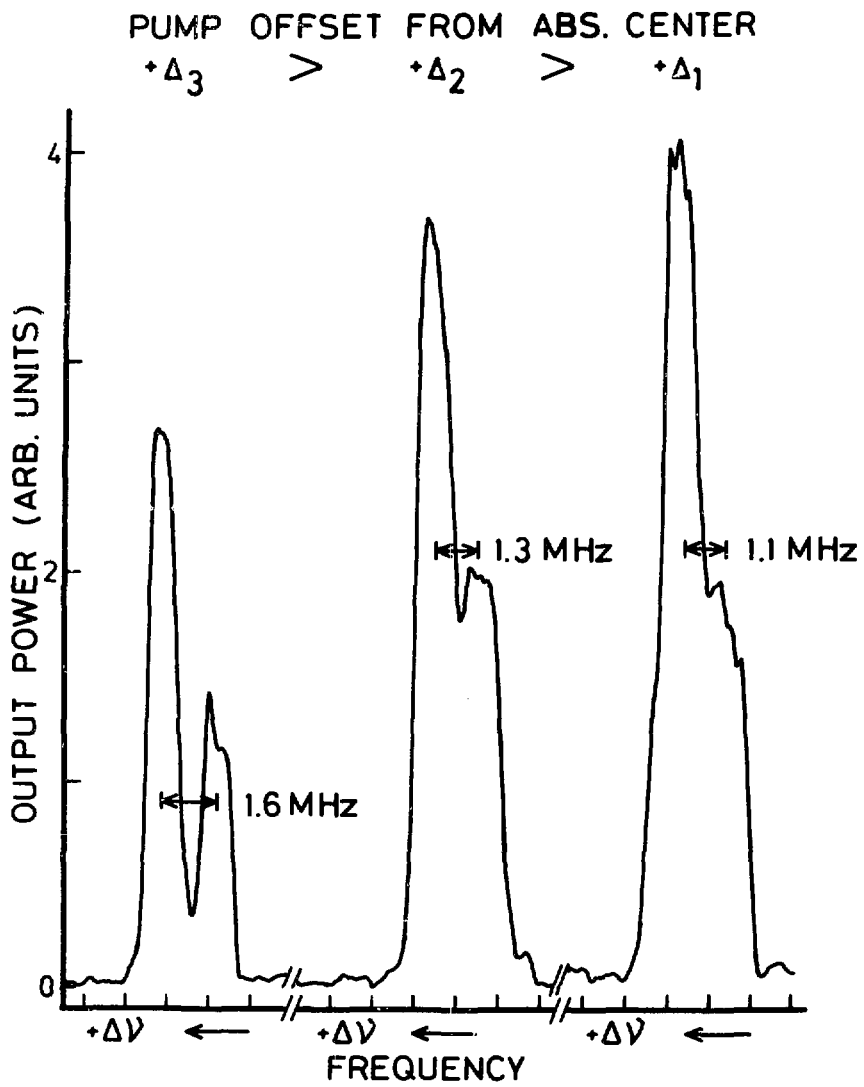


Fig.5

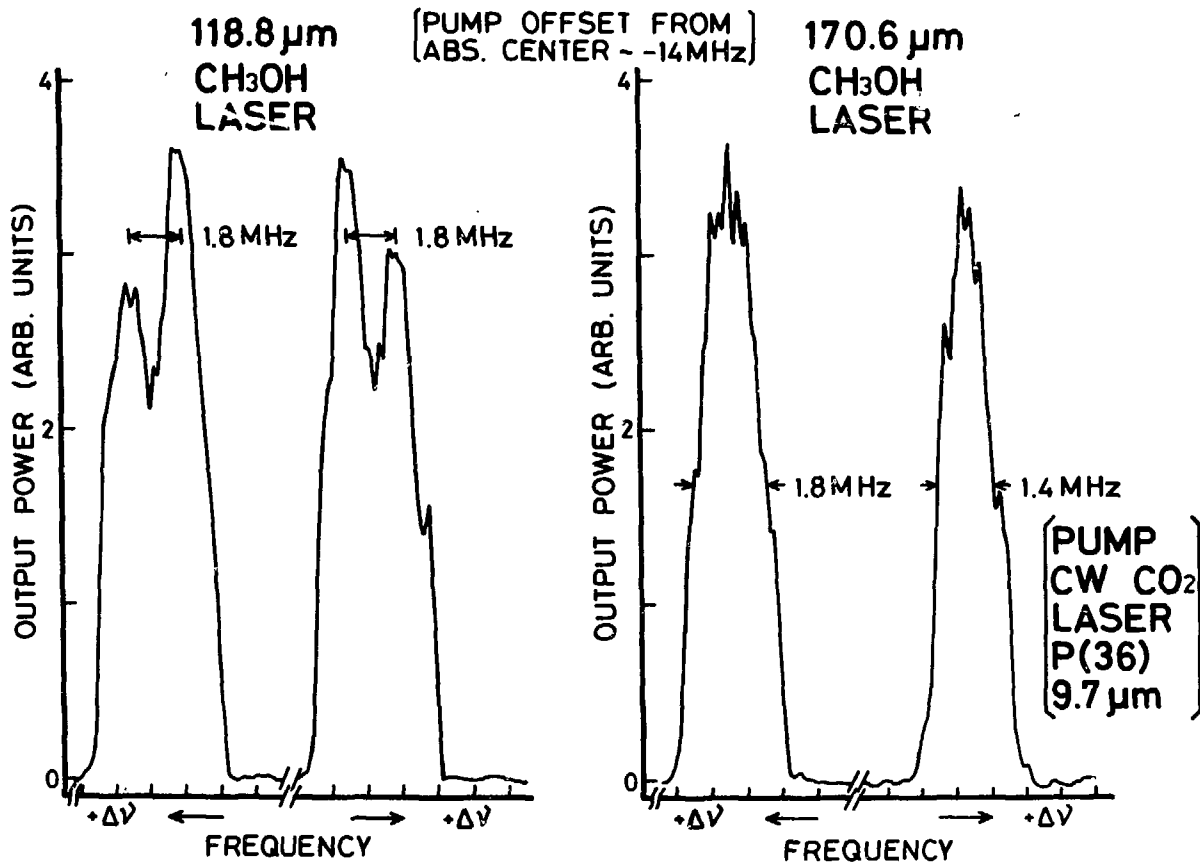


Fig.6

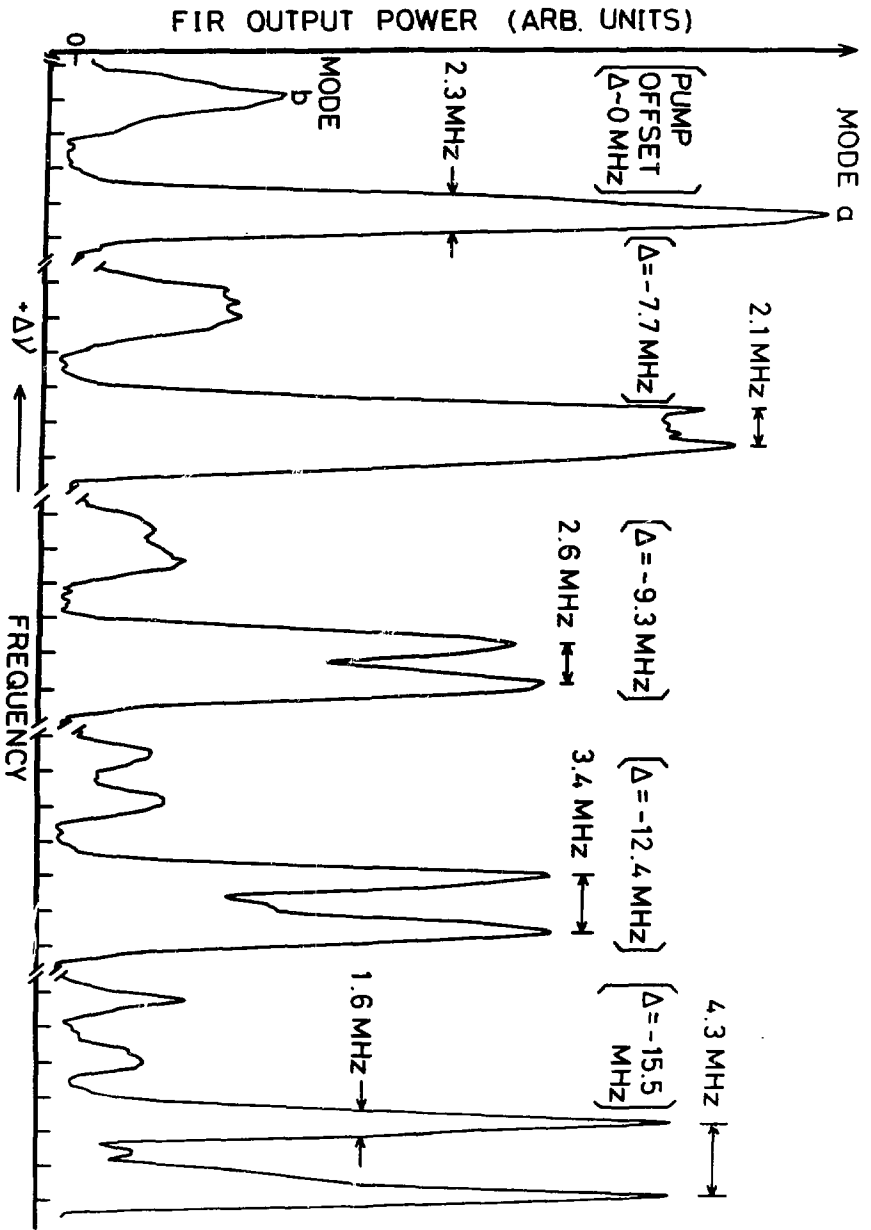


Fig. 7

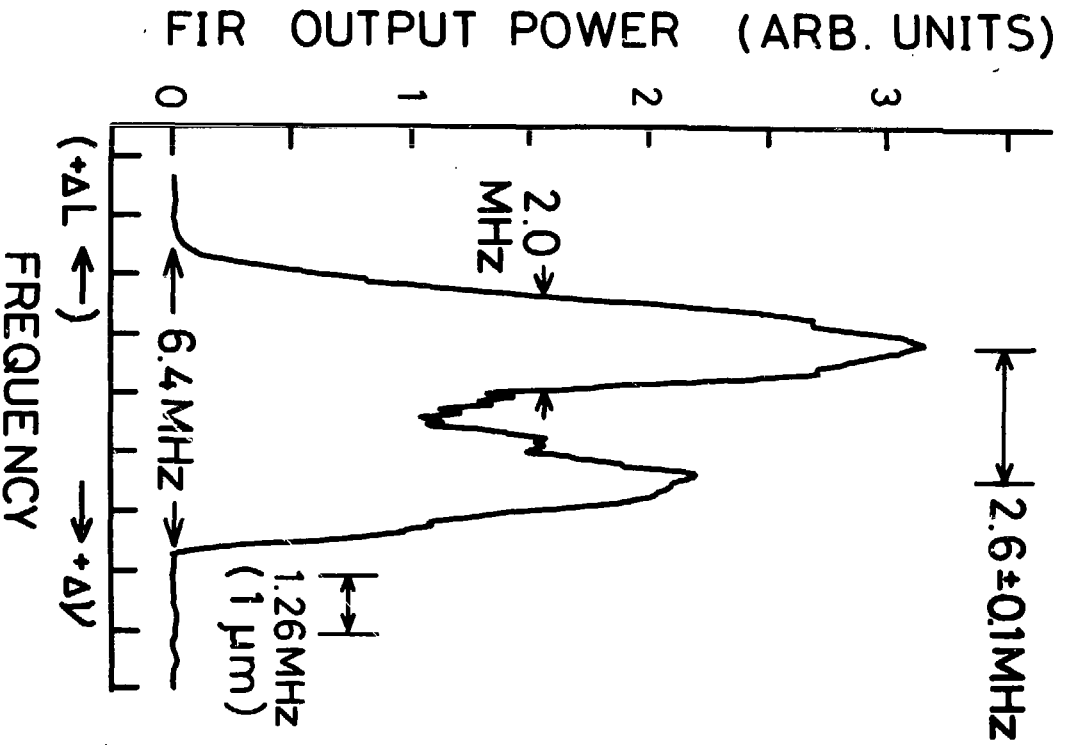


Fig. 8

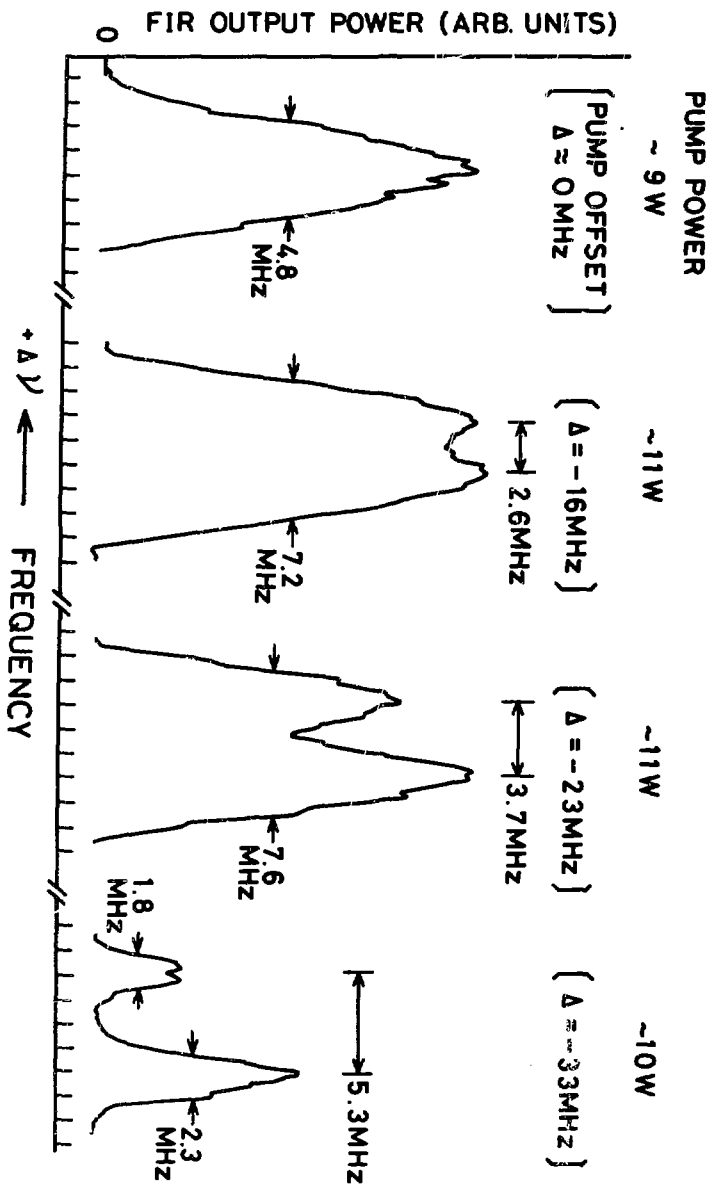


Fig. 9

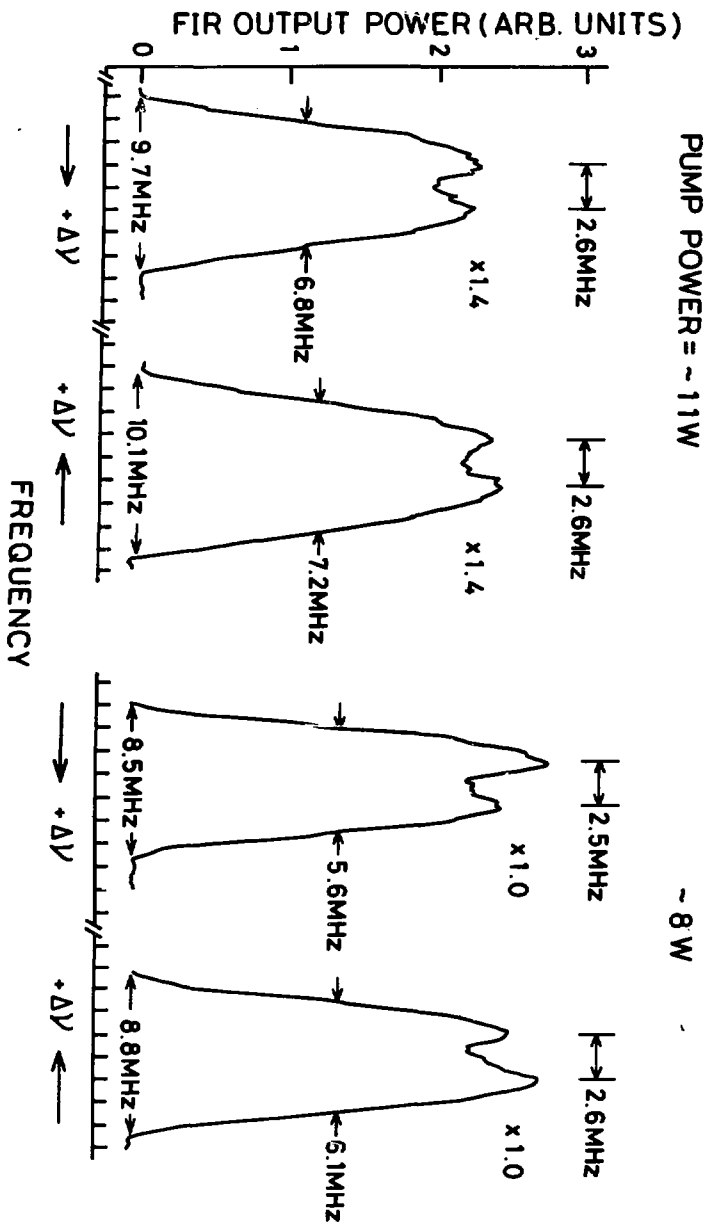


Fig. 10

CW 118.8 μm CH₃OH LASER

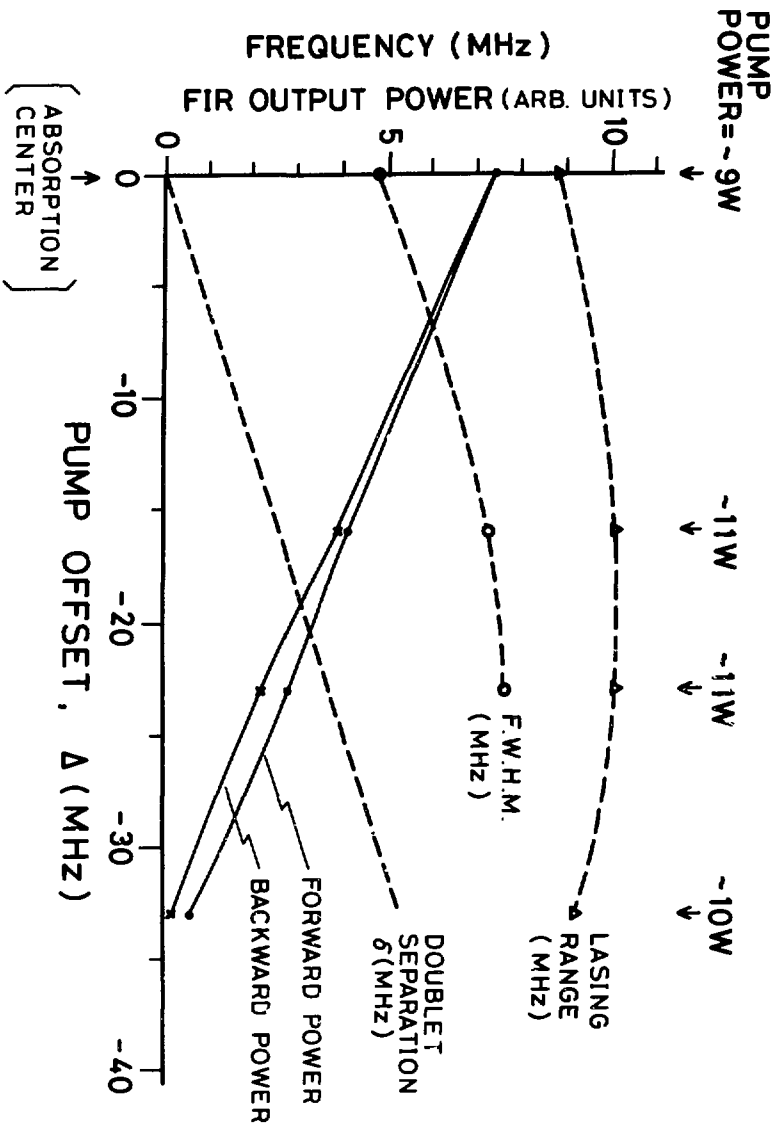


Fig. 11

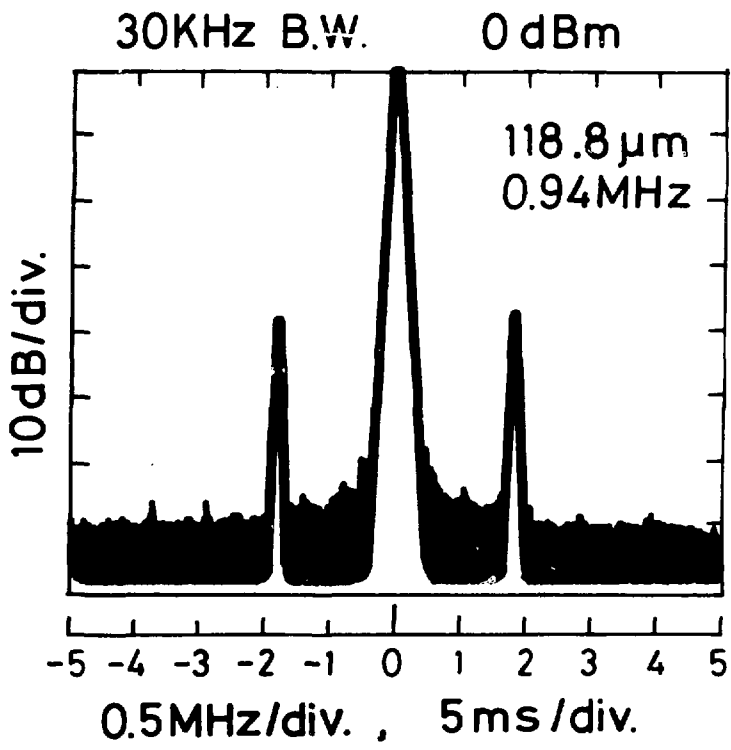


Fig.12

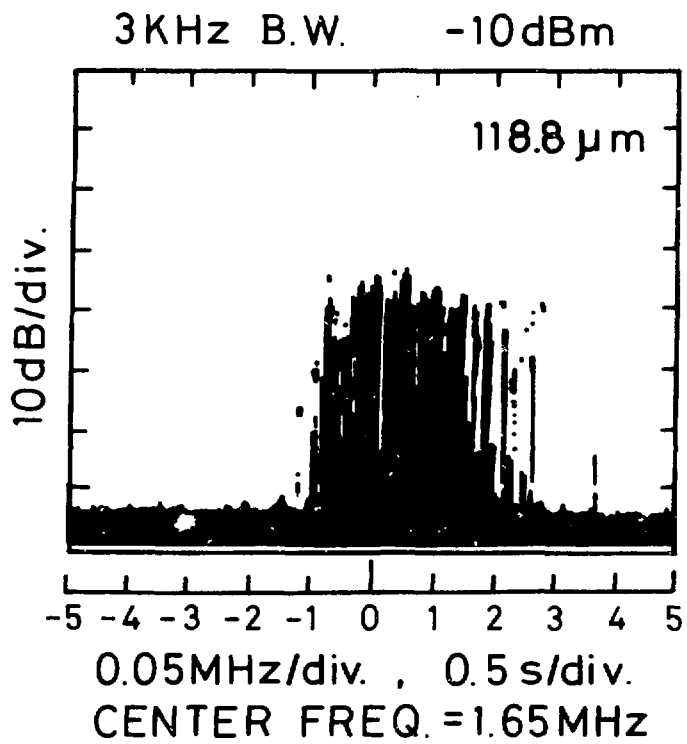


Fig.13

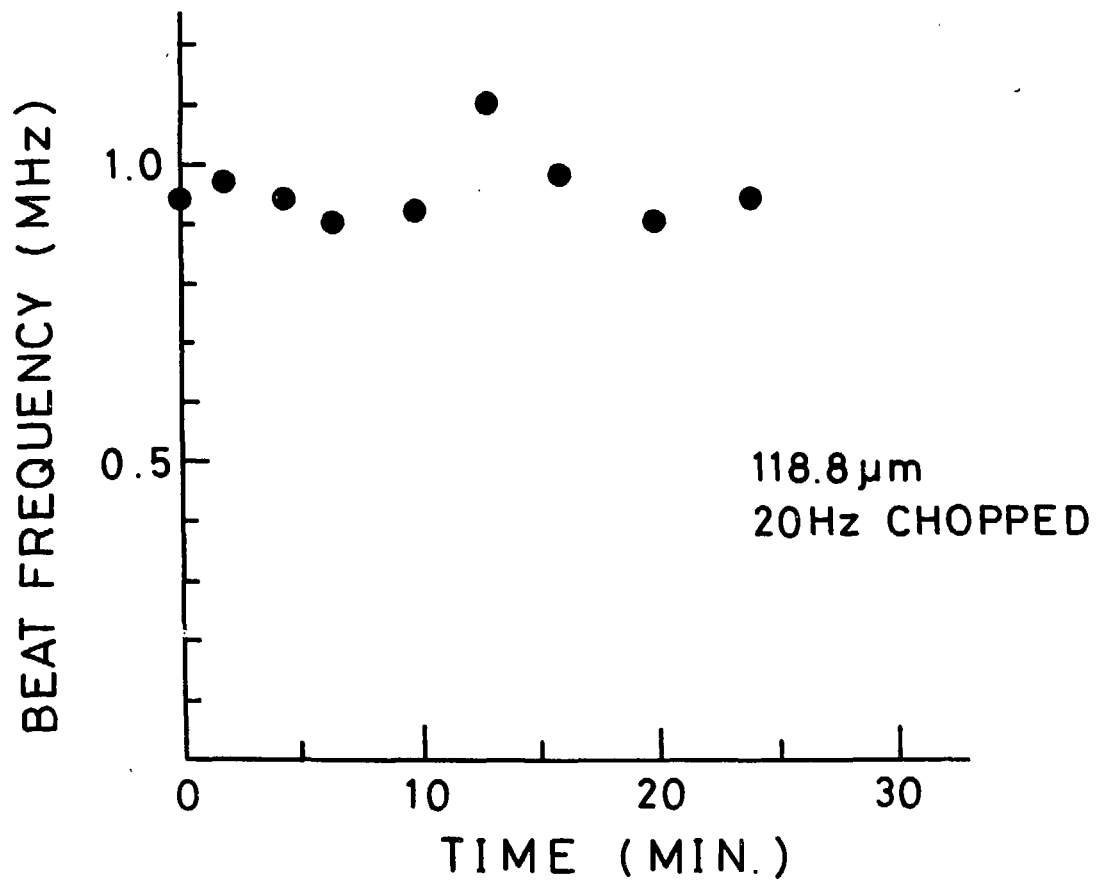


Fig.14