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ABSTRACT

The various types of mid-infrared optically pumped laser (OPML) are briefly reviewed, and the factors which determine the suitability of a molecular species for use in such a laser discussed.

Recent work on large-scale cryogenic cells for OPML's is described. These cells achieve reliable long-term operation in the 77-220K region with temperature stability and uniformity of ±1.5K. A novel heterodyne frequency locking system for use as an OPML pump source is also presented. This system offset locks a pulsed TEA to a CW source to better than ±500kHz.

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INTRODUCTION

A large number of optically pumped lasers operating in the far infrared (50μ-3mm) are now known, and the number of molecules which have been shown to lase in either pulsed or CW mode is still increasing rapidly. A very wide range of molecular types can be used; most compounds containing 7 or fewer atoms, having a fairly large permanent dipole moment and an infrared absorption band of moderate strength accessible to a CO_2 laser pump source, will work. In a few cases molecules as large as 10 atoms will lase (e.g. $OHCH_2CH_2OH$).

In strong contrast relatively few mid-infrared systems have been published, despite the intense interest in this spectral region. Fewer than twenty different chemical species have been described in the literature as providing mid-infrared (5-50μ) optically pumped laser (OPML) action, even when a variety of pump sources is considered. In the first part of this paper the various types of mid-infrared OPML are reviewed, and the constraints on the pump laser and molecular species which can be used are briefly discussed. In the second section recent work on large scale cryogenic cells and frequency-offset heterodyne locking techniques being used to scale up these lasers and improve their stability and efficiency is described.

PUMP LASERS

Pump laser requirements for OPML's are stringent. In general we require to resonantly pump a particular transition, and the source should ideally be line or continuously tuneable over the widest possible range, with a line width not exceeding that of the transition to be pumped.

The best chance of obtaining laser action in a new system is under conditions for maximum gain. As the OPML gas pressure is increased the gain rises linearly whilst the line remains Doppler broadened, becoming constant in the Lorentzian regime, which for these systems typically starts at 1-5 Torr. This in turn determines the best choice of pump pulse length, since the best chance of obtaining laser action is under collision free conditions. The pump pulse should therefore be $\leqslant$ 100ns. The pump pulse energy required depends strongly on the pump bandwidth and the strength of the pump transition. For weak pump bands and wide bandwidths a few Joules are typically required, whilst in the most favourable cases a few mJ will suffice. CW OPML action has not so far been observed in the mid-infrared. HF, DF, HBr, CO, CO_2 and doubled CO_2 lasers have all been used to pump mid-infrared OPML's. Of these the TEA CO_2 laser is by far the most important. It is the cheapest and most simple of these lasers, readily commercially available or home-made, and provides the widest range of single line tuned outputs with only marginal reduction in output energy when line tuned. Isotopic carbon and oxygen are increasingly used in closed cycle TEA CO_2 lasers, and the availability of sequence band lines still further extends the range of available pump lines. When fully developed the high pressure, continuously tunable CO_2 laser will remove the need to rely on chance line coincidence.

In contrast the hydrogen halide and CO lasers suffer severe reduction in total output energy on line tuning due to their cascade nature, and for similar sized systems provide much lower intensities. Whilst frequency doubled CO_2 lasers are a very attractive pump source the absence of commercially available, efficient doubling crystals such as $CdGeAs_2$ has greatly restricted their use. The TEA CO_2 laser gain switched spike of 50-150ns duration is ideal for pumping OPML's, and the rest of this paper will consider only CO_2 pumped systems.

The output bandwidth of a typical CO_2 TEA laser is 1.5GHz, much wider than the OPML pump transition. This is useful for survey purposes, but much improved efficiency can be obtained with line narrowed, resonantly tuned operation of the pump laser.

OPML SYSTEMS

The various major types of mid-infrared OPML are shown in Fig. 1 (Letokhov, 1972). The special case of two photon pumped systems (e.g. NH_3, CH_3F) is omitted.

In the 'light rotor laser' a light molecule with large rotational constants is excited on a fundamental, and laser action occurs back to a higher lying rotational level of the ground state. Actual inversion (as opposed to Raman laser action) can only be obtained at an absolute temperature T (kelvin) if the separation of the initial and final states is $\geqslant 0.6kT$, and the scheme is only applicable to very light molecules such as NH_3. The NH_3 12.8μ laser operating in this manner is the most powerful and efficient mid-infrared laser. Unfortunately the prospects for new systems of this type are severely limited by the restriction to very light molecules.

The material may alternatively be excited on a fundamental and lase on a difference band. This scheme shares with the light rotor laser the advantage of strong pump absorption, but suffers from low gain on the difference band laser transition.

Since difference band with sufficient strength to permit laser build up within the inversion lifetime are rare, again the prospects for new systems are limited.

The most versatile scheme utilizes excitation of a combination band with laser action terminating in one of the combination band component vibrations. The pump absorption is weak, typically only a few percent per metre, but the laser gain is high and superfluorescence easily obtained. This system has provided most of the new mid-infrared OPML's. The currently published systems of this type are shown in Table 1.

MOLECULES FOR COMBINATION BAND PUMPED OPML'S

Many criteria affect the suitability of a molecule for use in an OPML. Evidently the possession of an appropriate pair of energy levels whose combination band is symmetry allowed is essential. Many of the other factors may be considered in a very simple manner which is still adequate for guidance in choosing systems to be screened for OPML action.

i. Laser transition strength. The inversion is effectively destroyed at approximately the gas kinetic rate by R-R collisional transfer. For pressures $\sim$ 1 Torr, and assuming a short pump pulse this lifetime is 100ns.

To obtain build up from noise we require

$$\alpha c \tau \geq 30$$

where α is the laser gain, τ the inversion lifetime and c the speed of light. Hence we require $\alpha \geqslant 1\%\ \mathrm{cm}^{-1}$, corresponding to a moderate strength infrared band.

ii. Pump transition strength. To just achieve inversion we require

$$\frac{\Delta\nu_A}{\Delta\nu_p}\,\frac{I\alpha_p}{h\nu} = \exp(-E/kT)\ Nd/Q$$

where I is the pump intensity, (J cm^{-2}), α_p the pump absorption coefficient, ν the pump frequency, E the lower laser level energy, Q the total partition function, d the lower laser level degeneracy (we assume d $>>$ 1) and $\Delta\nu_p$, $\Delta\nu_A$ the pump laser and absorption line widths respectively. The other symbols have their usual meaning. Taking typical conditions of 1 Torr, 200K and a 400 cm^{-1} lower level we have

$$I\,\alpha_p \geq 4 \times 10^{-5}\ (\Delta\nu_p/\Delta\nu_A).(d/Q)$$

Since d/Q is typically $<<$0.1 and the ratio of line widths 25, and I$\sim$1 is easily achieved with a CO_2 TEA laser, even extremely weak pump absorptions will suffice. Such weak combination bands are often not reported in the literature, and are easily obscured by overlapping line wings, impurities, etc. Weak pump bands clearly demand long path lengths for efficient operation, placing constraints on the pump laser divergence and making OPML cryogenic cell design cumbersome.

iii. Overlapping absorption. Lasers of this type are automatically overlapped by the corresponding absorbing transition from the ground state. The rotational structure of the transition must be resolved at Doppler limited resolution for laser action to be possible. The total overlapping line density is greatly increased if other low lying modes (typically distortional or torsional) give rise to numerous overlapping hot bands.

iv. Isotopic composition. Mono-isotopic species have the best chance of OPML action. In general only one isotopic species is pumped at a time, the others giving rise to overlapping absorptions and rapid, resonant relaxation processes. For elements with minor isotopes with abundances of a few percent such as C, O, N and S the effect is negligible. However for elements such as Cl, Br where two or more isotopes are present in comparable abundance laser action is unlikely in unenriched material, rendering screening of such compounds very costly. The effect is particularly serious if more than one atom of the multi-isotopic element is present in the molecule. There is a noticeable preponderance of compounds having little 'isotopic confusion' in Table 1.

v. Partition function. Under collision free conditions the extractable energy density depends directly on the total partition function, since this partly determines the number of molecules available in the lower level of the pump transition. The vibrational partition function is dominated by the lowest lying modes and their degeneracy, whilst the rotational partition function depends on the molecular constants and symmetry. Low lying, degenerate modes again adversely affect the OPML.

The very low value of the rotational partition function in the light, linear molecule C_2D_2 (~200) as compared to most other OPML's (e.g. ~ 3×10^4 for $FClO_3$) partly accounts for the fact that this molecule is the most powerful OPML of this type.

vi. Relaxation rates. Aside from inducing overlapping hot band absorptions and increasing the vibrational partition function, low lying modes correlate with rapid V-T relaxation rates (the Slater-Lambert law) which in extreme cases can be as fast as R-R rates. The R-R rate is usually within a factor of 2-3 of gas kinetic for most molecules, and so cannot be strongly influenced.

vii. Chemical and physical properties. Virtually all the adverse effects are reduced at low temperatures, and only the C_2D_2 OPML works well at 300K. The vapour pressure curve for the materials is thus of paramount importance. There are only a limited number of high damage threshold, mid-infrared transparent materials, and compatibility with these is a problem in some cases (e.g. SiF_4, NSF).

viii. Competing transitions. If both components of the combination band are infrared active it may be necessary to suppress the component which is not required. Since many of the molecules have large permanent dipole moments and are known FIR lasers (e.g. OCS, CH_3CCH)

interaction with FIR laser transitions can occur in both beneficial and deleterious manners.

The qualitative criteria discussed above agree well with the properties of the molecules listed in Table 1, and in general those molecules which best satisfy these criteria are the most efficient lasers. In further support of the importance of these factors we have found that natural isotopic composition CBr_2F_2, $CBrClF_2$, $SiHCl_3$ and $SiCl_4$ do not operate as OPML's despite possession of appropriate energy levels and observable combination bands. These molecules have over dense rotational structure, unwanted low lying modes and multiple isotopic forms which combine to prevent laser action.

LARGE SCALE CRYOGENIC CELLS

Mid-infrared OPML's require cryogenic cells of several metres length and 10-150mm aperture operating in the 100-250K region. Most systems have used cells cooled by the gaseous boil-off from liquid nitrogen. This uses the coolant inefficiently, and leads to slow thermal response, limited operating temperature range, poor thermal uniformity and restricted scaling capability. Fig. 2 shows the schematic design of a cell which achieves 1.5K stability and uniformity over 8 hour periods, has rapid thermal response and is easily scaled. The current cells are 2.5m long and 40mm bore, but this can be scaled to 150mm. The OPML gas is contained in a heavy wall, high conductivity copper pipe on which is wound a segmented, PTFE insulated heater. The tube is enclosed in an annular liquid nitrogen tank, and partially insulated from it by a nitrogen gas purged vermiculite layer. By adjusting the total heater power and power profile an operating range of 77K to 220K is readily achieved.

HETERODYNE FREQUENCY OFFSET LOCKING

Greatly increased power and efficiency can be obtained from the OPML by narrowing the CO_2 laser bandwidth to match the OPML.

A convenient way to achieve this is by injection locking the main TEA CO_2 laser with a frequency stabilized, tuneable source. A miniature TEA CO_2 laser operating TEM_{00} mode and with a short, 150mm cavity has been developed for this purpose. A tuning range of 250MHz about line centre is achieved using a piezo-electric mirror drive.

Fig. 3 shows the electronic system we have utilized to frequency stabilize this laser. The TEA laser pulse is heterodyned with a frequency stabilized CW CO_2 laser on a wideband room temperature MCT detector. The heterodyne signal is filtered and amplified, and applied to the circuit shown schematically in Fig 3. A limiting amplifier removes amplitude variations, and the limited r.f. pulse is applied to a cosine frequency discriminator constructed from a 3dB power divider, cable phase delay and a double balanced mixer. The centre frequency is simply changed by altering the phase delay (cable length). The output of the frequency discriminator provides a direct measure of the pulse chirp, but for frequency lock purposes is sample-held, integrated and drives a high voltage amplifier which in turn drives a piezo-electric tube, so completing the loop.

Fig. 4 shows an open loop opto-acoustic scan of the $^{P}R_4(29)$ line of $^{12}CH_3{}^{81}Br$ -103MHz from $^{12}C^{16}O_2$ 10P18, with a simultaneous scan of the discriminator output voltage, which has been adjusted to provide a zero crossing at the peak of the CH_3Br line. Fig. 5 shows the open loop frequency of the miniature laser, deliberately perturbed by air drafts. Closing the loop dramatically improves the stability, and more recent results have shown periods in excess of 3 hours with the frequency averaged over a 10 second time constant remaining within 500kHz of the desired value. It must be emphasized that no 'dither' is employed, and in principle shot-by-shot correction can be used. Also continuous, locked tuning can be achieved by replacing the phase delay cable with an adjustable phase shifter. It is anticipated that this stabilized laser will form an extremely convenient, high repetition rate (up to 100Hz) source for injection locked control of OPML pump lasers.

CONCLUSIONS

Mid-infrared OPML's are by far the most convenient laser source in this wavelength region. The simplicity and low cost of these systems make them very attractive as wavelength convertors for use with CO_2 TEA lasers. The physics of these systems is still not completely understood, and much interesting work remains to be done on both their physics and technology.

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TABLE 1

MID-IR OPTICALLY PUMPED LASERS

CO_2 LASER PUMPED VIA A COMBINATION BAND

Pump Gas	Band μ	Laser Band μ		
OCS	9	19	020 - 010	Schlossberg 1975
$^{12}CF_4$	9	16	$\nu_2 + \nu_4 - \nu_2$	Tiee 1977
$^{13}CF_4$				
$^{14}CF_4$				
NOCL	10	16	011 - 001	Tiee 1977
NSF	10	16	011 - 001	Fischer 1980
CF_3I	9	13-14	$\nu_2 + \nu_3 - \nu_3$	Tiee 1978
CH_3CCH	10	16	$\nu_9 + \nu_{10} - \nu_{10}$	Fischer 1981
$^{12}C_2D_2$	9	17-20	$\nu_4 + \nu_5 - \nu_4$	Rutt 1978
$^{13}C_2D_2$				
SiF_4	9	25	$(2\nu_4 + \nu_2)-(\nu_4 + \nu_2)$	Green 1979
$FClO_3$	10	16-18	$\nu_5 + \nu_6 - \nu_6$	Rutt 1980

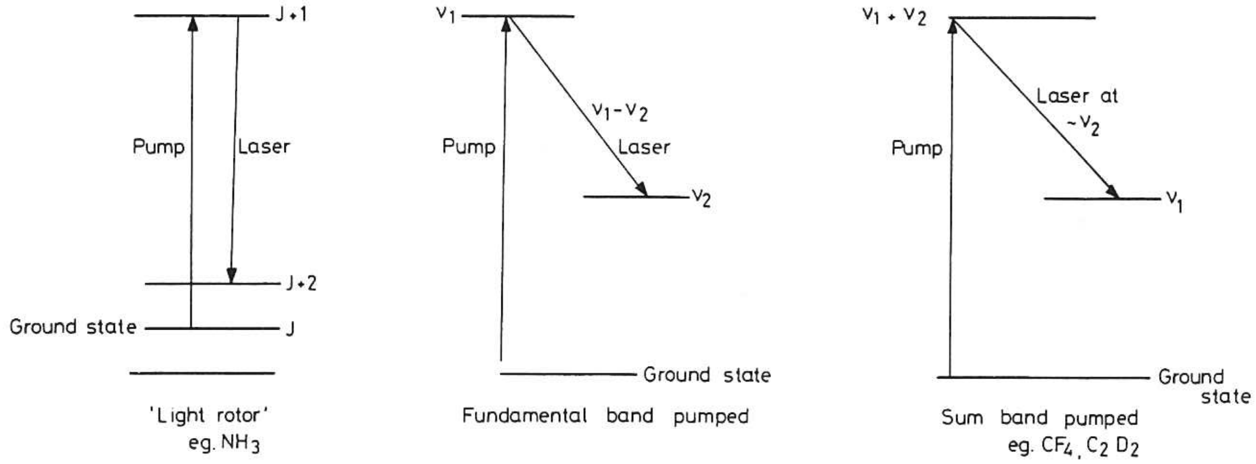


Fig.1 MID-IR optically pumped lasers.

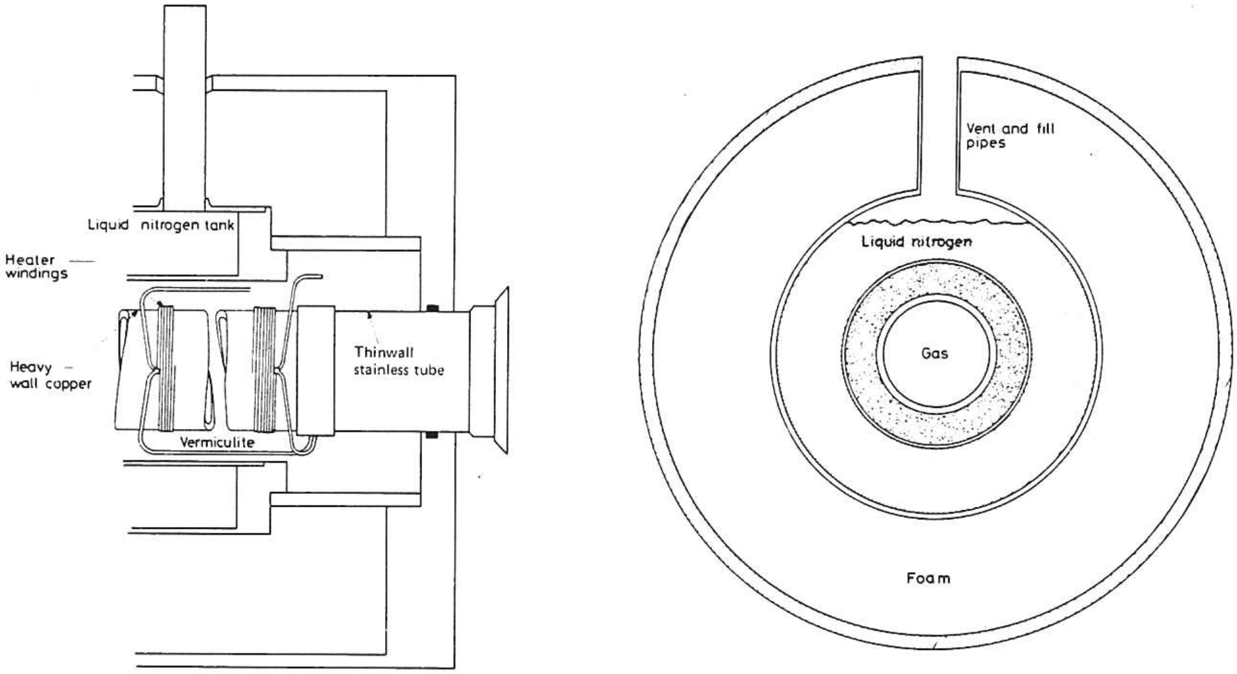


Fig.2 Large scale cryogenic cell.

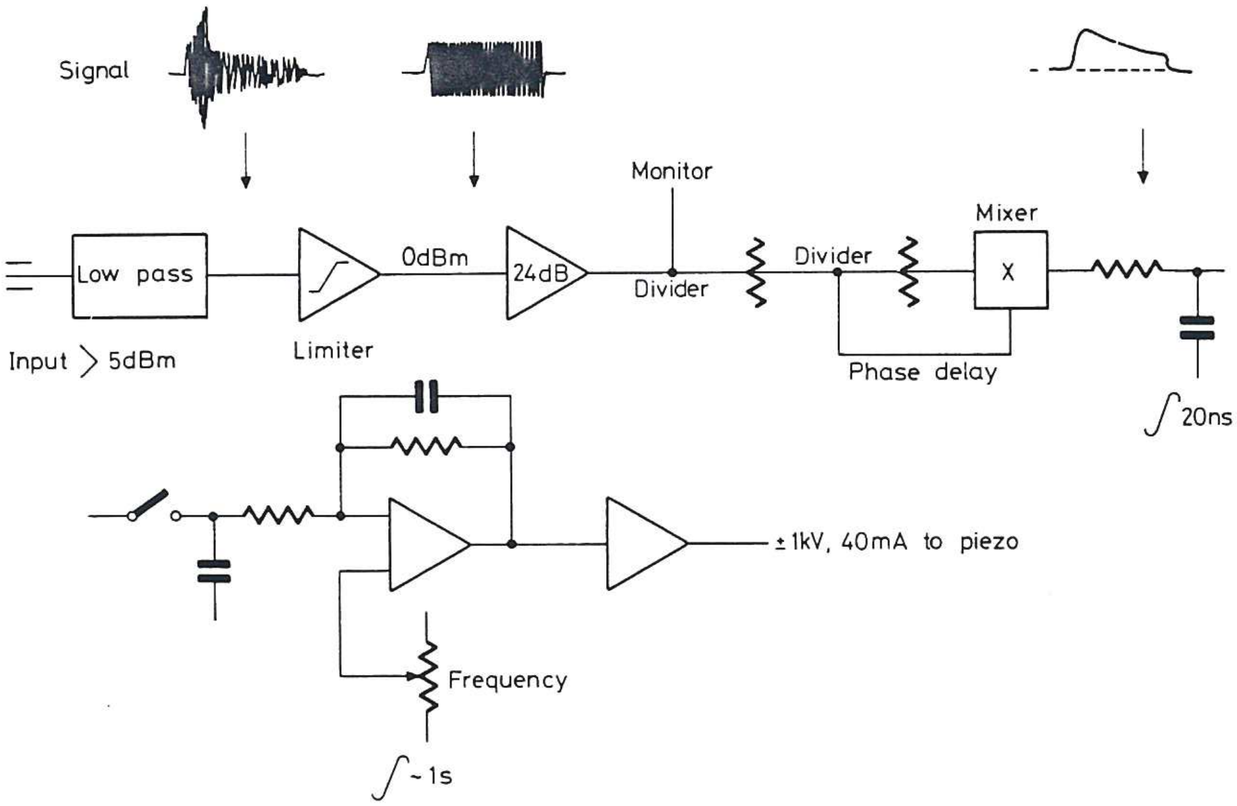
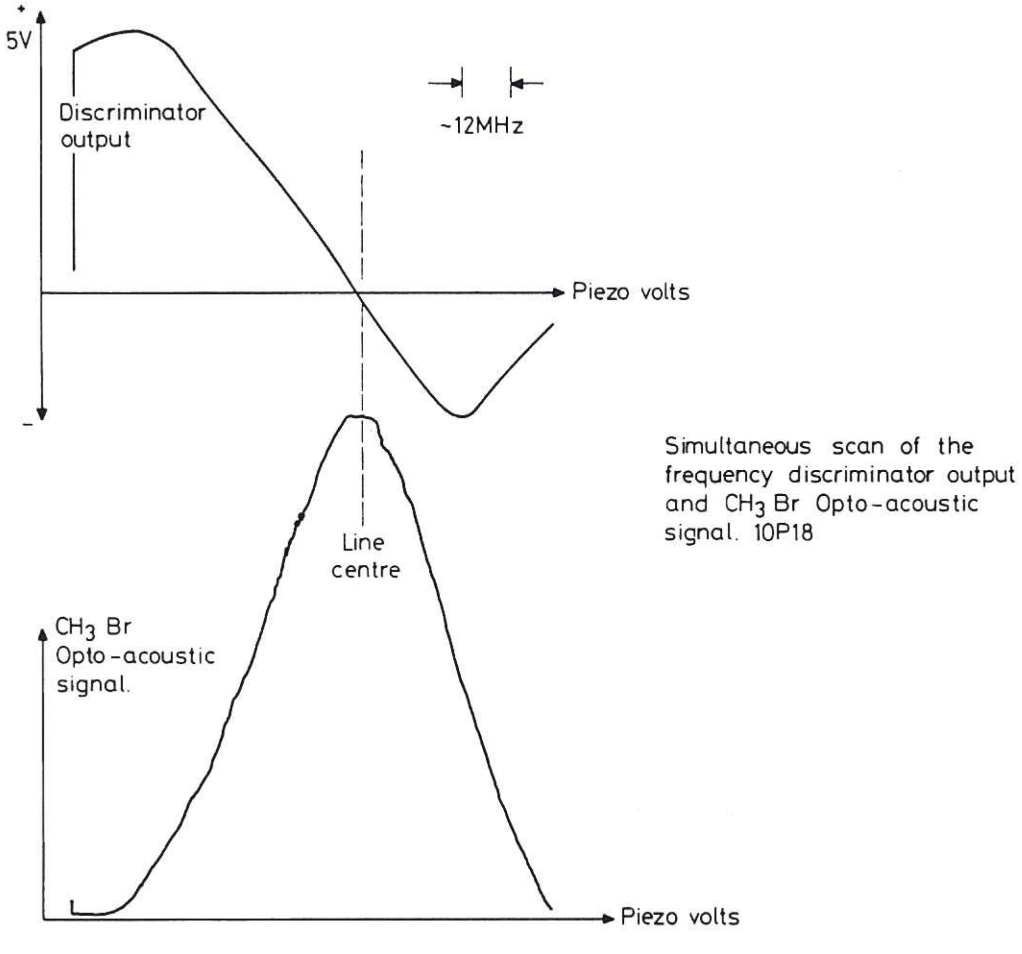


Fig.3 Heterodyne signal processing.



Simultaneous scan of the frequency discriminator output and CH_3 Br Opto-acoustic signal. 10P18

Fig.4 Frequent discriminator response.

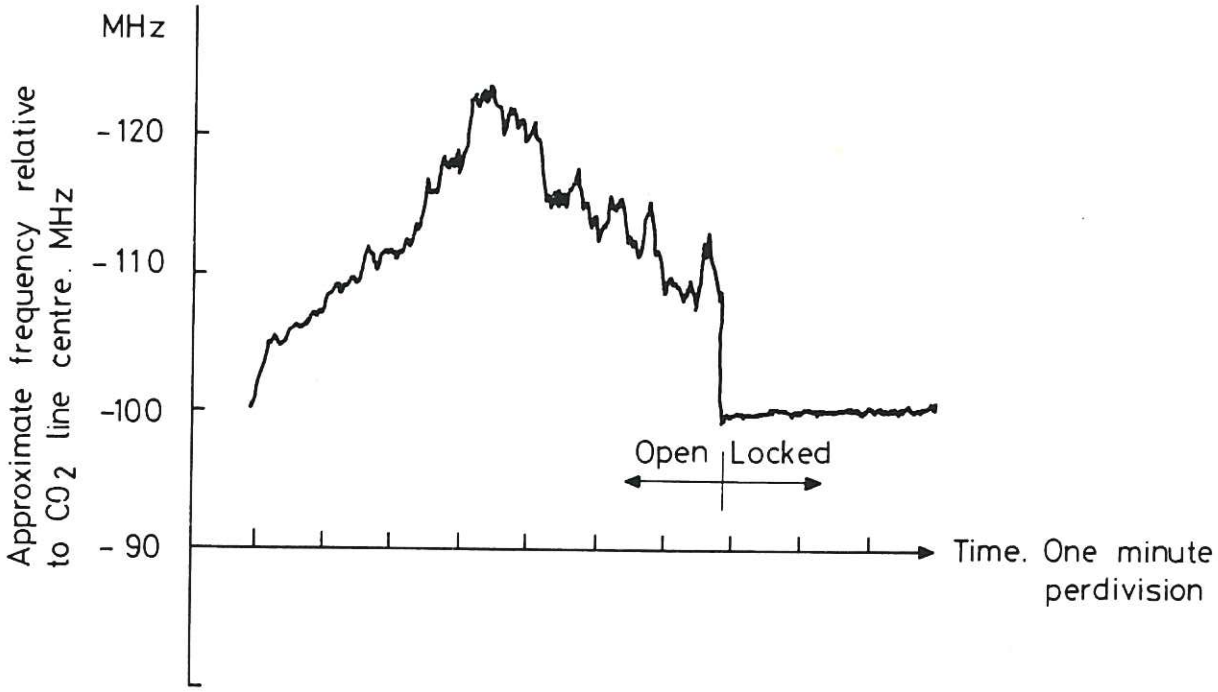


Fig.5 Closed loop frequency stability.