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SELF FOCUSING AND DEFOCUSING OF PUMP RADIATION IN A FAR-INFRARED LASER

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ABSTRACT

For optically-pumped FIR lasers the frequency of the pump beam is in close coincidence with an absorbing transition of the active medium. Saturable absorption results in an intensity-dependent local refractive-index, leading to self focusing or defocusing. The latter effect has been demonstrated experimentally in CH_3F, wherein focusing lengths of the order of several metres were measured, in agreement with theoretical predictions. This phenomenon has important consequences in the design of high-power FIR amplifiers.

1. INTRODUCTION

Optically-pumped pulsed FIR lasers are currently under development for plasma diagnostics. In general, narrow-line output pulses in excess of 1 MW peak power are necessary [1,2]. This requires amplifier chains several meters long. In the case of axial pumping, the pump beam propagates in the laser medium over long distances with a frequency close to an absorbing transition. For optimum efficiency, one aims to operate these lasers in the saturated regime. This not only leads to an intensity-dependent absorption coefficient, but also to an intensity-dependent refractive index and hence to self focusing or defocusing. The former would result in nonuniform excitation of the laser gas and could even lead to gas breakdown, whereas in the latter case pump radiation could be lost to the walls of the amplifier tube. If one is aware of these problems they are relatively easy to overcome by adjustment of the divergence of the beam before it enters the amplifier. In this paper we give a general relationship between characteristic self focusing or defocusing lengths and the absorption coefficient, and present experimental results demonstrating self defocusing in methyl fluoride (CH_3F).

2. SELF FOCUSING IN THE VICINITY OF AN ABSORBING TRANSITION

The possibility of self focusing of a laser beam due to the intensity-dependent dispersion of a resonance line has been discussed theoretically by Javan and Kelly in 1966 (3). Grischkowsky (4) reported experimental observations of the phenomenon with ruby laser light in potassium vapor.

We start from the equations relating the refractive index n and the absorption coefficient α to the complex dielectric constant ε and the real and imaginary susceptibilities χ' and χ'', respectively

(1)
$$n = \mathrm{Re}(\varepsilon)^{1/2} = \{1/2\,(1+\chi') + 1/2\,(1+2\chi'+\chi'^2+\chi''^2)^{1/2}\}^{1/2}$$
$$\alpha/(2k) = \mathrm{Im}(\varepsilon)^{1/2} = \{-1/2\,(1+\chi') + 1/2\,(1+2\chi'+\chi'^2+\chi''^2)^{1/2}\}^{1/2}$$

where $k = 2\pi/\lambda$ and the refractive index far from resonance has been set to unity. For a two level system the susceptibility due to the presence of the homogeneously-broadened transition can be expressed in the following way (5)

(2)
$$\chi' = -\chi_0\,\delta/(1+\delta^2)/(1+I/I_{sat})$$
$$\chi'' = \chi_0/(1+\delta^2)/(1+I/I_{sat})$$ *)

*) For a monochromatic beam
$$\chi'' = \chi_0/(1+\delta^2)\{(1+\delta^2)/(1+\delta^2+I_\nu/I_{sat}(\nu))\}.$$
For a broad-band laser beam the intensity-dependent term in curly brackets, suitably convoluted with the lineshape of the absorbing transition, has to be integrated over the bandwidth of the beam. In writing the term as in (2) we have taken this into account and I_{sat} is defined accordingly.

where δ is the frequency offset from the center of the absorbing transition in terms of full line width at half maximum and I is the beam intensity. The saturation intensity I_{sat} is defined as the beam intensity for which the absorption coefficient is reduced to half its low intensity value for zero offset. χ_0 is the resonance value of χ'' at negligible beam intensity. Combining equations (1) and (2) yields the dispersion relation and absorption coefficient near the transition frequency. Due to the intensity dependence of the refractive index, different ray bundles in the beam cross-section propagate with different phase velocities, which affects the beam divergence. On the high frequency side of the absorbing transition the refractive index n increases with increasing intensity; hence an initially parallel beam will self focus. The reverse is true on the low frequency side.

The procedure illustrated in fig. 1 can be used to estimate the beam divergence of a parallel beam due to self defocusing. The caustic of a Gaussian beam in a medium with nonlinear refractive index can be described by [6]

$$r^2 = \theta^2 z^2 + r_0^2 \tag{3}$$

with axial distance z, beam radius r, beam waist r_0, and far-field divergence half-angle θ. From this equation the radius of curvature of the caustic at the beam waist is easily derived

$$R_0 = r_0 / \theta^2.$$

Taking the point C of fig. 1 as the half-intensity point, we require that the optical path lengths AB and CD be equal,

$$\overline{AB} \cdot n(I) = \overline{CD} \cdot n(I/2)$$

with
$$\overline{CD} = R_0 \cdot \Delta\varphi$$

$$\overline{AB} = \overline{AE} + \rho\,(1 - \cos\Delta\varphi)\,.$$

For small $\Delta\varphi$, $\overline{AE} \approx \overline{CD}$, $\rho \approx r_0/\Delta\varphi$, and $\cos\Delta\varphi \approx 1 - {}^1\!/_2\,(\Delta\varphi)^2$, so that one obtains

$$\theta = 2^{1/2}\,\{n(I/2)/n(I) - 1\}^{1/2}\,. \tag{4}$$

It can be shown that the same angle is obtained for a converging beam, if the ratio n(I/2)/n(I) is inverted.

The divergence of a beam due to the presence of a saturable absorbing transition can now be computed from equations (1), (2) and (4). Usually $\chi_0 \ll 1$, which allows certain approximations, thereby simplifying the calculations. One finds

$$\begin{aligned} \alpha &= k\chi'' = k\chi_0\,(1+\delta^2)^{-1}\,(1 + I/I_{sat})^{-1} \\ \theta &= \pm\{\chi_0|\delta|\,(1+\delta^2)^{-1}\,{}^1\!/_2\, I/I_{sat}\,[(1 + I/I_{sat})\cdot \\ &\qquad (1 + {}^1\!/_2\, I/I_{sat})]^{-1}\}^{1/2}\,. \end{aligned} \tag{5}$$

Fig. 2 shows plots of the far-field divergence angle and the absorption coefficient versus offset from the line frequency, with the ratio of beam intensity to saturation intensity as parameter. Self-focusing occurs on the high-frequency side of the resonance and self-defocusing on the low frequency side (not shown). Within the wide range of intensities shown, and for frequency offsets of up to ten absorption linewidths, there is surprisingly little variation in the self-focusing effect.

Experimentally it is very simple to measure the absorption coefficient, but χ_o is not usually known. Therefore, it is convenient to have a relationship between absorption coefficient and divergence angle. One finds from equation (5)

$$\theta = \pm (\alpha / k)^{1/2} \{ |\delta| \; ^1/_2 \; I / I_{sat} / (1 + {}^1/_2 \; I / I_{sat}) \}^{1/2}. \tag{6}$$

In the saturated regime the angle Θ is an insensitive function of intensity and varies only with the square root of the frequency offset. Thus a reasonable estimate of self focusing can be made even if the precise frequency of the absorbing transition is unknown.

3. EXPERIMENT

To demonstrate self defocusing of a CO_2 laser beam in methyl fluoride, the beam was passed through an absorption cell containing the gas (fig. 3) and the position of an air spark, produced by focusing the attenuated beam with a concave mirror, was observed. Variations of the beam divergence resulted in axial displacements of the air spark. The doubledischarge CO_2 laser was able to produce output pulses of up to 20 J. For the current experiment it was tuned to the P(20) transition at 9.55 μm with a diffraction grating. This transition is in close coincidence with the Q (12,2) ($\nu_3 = 0 \rightarrow 1$, $J = 12 \rightarrow 12$, $K = 2 \rightarrow 2$) absorption transition in CH_3F. With an aperture of 1 cm diameter in the resonator, pulse energies of 1.6 J were measured behind the entrance window of the 135 cm long absorption cell. About 25 % of the energy was contained in a short pulse of 40 nsec duration and the rest in a 1.5 μsec tail. Air sparks were produced by means of a curved mirror of 50 cm focal length.

The effect of varying the beam intensity on the position of the sparks was carefully studied. It was found necessary to operate close to the breakdown threshold since the sparks were displaced towards the focusing mirror at higher intensities. The positions of a large number of air sparks were recorded photographically with and without CH_3F in the cell. To avoid ambiguities, records which showed several sparks were discarded. We believe that multiple sparks were due to dust particles in the air. Usually only one spark showed the characteristic elongated shape, whereas the others were spherical. Control measurements with laser pulses at 10.6 μm were also carried out. At 9.55 μm an average distance between mirror and spark of 53.8 cm was measured with the cell evacuated and 57.7 cm with 5 torr of CH_3F, with standard deviations of 1.0 cm and 0.65 cm, respectively. No significant difference was observed with 10.6 μm radiation between using an evacuated cell and a cell with 5 torr CH_3F.

To interpret the results, a t - test [7] was performed on the sample means and data distribution. The test demonstrated, with a confidence level exceeding 99 %, that at 9.55 μm the two sets are from different populations - i.e. the effect of the CH_3F on the position of the focus is significant. At 10.6 μm there is no evidence of the CH_3F having a significant effect.

From the measured offset of 3.8 cm from the nominal focal length of 50 cm in the case of an empty cell, the divergence of the laser beam is obtained. Taking this into account and describing the propagation in CH_3F by means of equation (3) allows one to compute the effect due to the gas cell. We find that it is equivalent to the effect of a negative lens of 350 cm focal length positioned at a distance of 53 cm from the entrance window of the cell. Hence the divergence due to self defocusing is $\Theta = 1.4$ mrad.

4. DISCUSSION

The divergence can also be estimated from our theoretical considerations, knowing the absorption coefficient, saturation parameter and frequency offset. The low-intensity absorption coefficient at resonance, measured by Hodges and Tucker [8], is $\alpha\ (\Delta\nu = o,\ I = o) = 0.2\ cm^{-1}$ for 5 torr of CH_3F. From the same reference we obtain the saturation intensity: $I_{sat} = 0.15$ MW (5 torr). The beam intensity is obtained from our measured data. In order to compute the saturation parameter, it should be remembered that only the radiation intensity in the frequency band corresponding to the absorption width is relevant. At 5 torr the absorption width is 200 MHz [8], whereas the CO_2 line width is typically 2 - 3 GHz [9]. Taking this into account, we obtain a saturation parameter I/I_{sat} in the range 6-8.

Each frequency component is subject to a different amount of self focusing or defocusing. We are interested in the spectral part with the highest intensity. Originally this was the line center, but due to absorption a 200 MHz wide dip is formed in the frequency spectrum of the laser line, so that the intensity peaks are displaced from the absorption line center by 2 to 3 absorption line widths. It is now possible to compute the beam divergence by means of equation (5). One obtains $\Theta \approx 1$ mrad, in fair agreement with the experimental value.

The line centers of the absorbing transition in CH_3F and the CO_2 P(20) line are separated by about 40 MHz [8]. From their relative positions self defocusing of the P(20) radiation is predicted, in agreement with our observations. However, in comparison with the laser linewidth this displacement is so small that a considerable fraction of the total laser power is expected to be on the high-frequency side of the transition. A second less-intense spark, due to self focusing, would be expected on the opposite side of the mirror focus. This was not observed.

A possible explanation for the missing air-spark is the presence of other absorbing transitions on the high-frequency side of the CH_3F absorption line under consideration. For example, sublevels with higher quantum numbers K of the same $J = 12$, $\nu_3 = 0 \rightarrow 1$ transition could be responsible. Also, there is a $\Delta J = -1$ transition on the high-frequency side of the main absorption line, separated from it by $\sim$ 1.6 GHz. Furthermore there is coupling between different frequency components, not accounted for in our simple model. A detailed numerical analysis, as in [10], should reveal the reason for self defocusing only, under our experimental conditions.

Similar experiments have been performed with the CO_2 P(22) line at 9.57 μm in 10 torr of CH_3F. This line is on the low frequency side of two close absorption lines ($J = 17 \rightarrow 17$ and $J = 1 \rightarrow 2$ for $\nu_3 = 0 \rightarrow 1$), both separated from the laser line by about 4 GHz. Again self defocusing is observed with a divergence of the order of 1 mrad.

The treatment previousely outlined can, of course, be applied to other FIR laser materials, permitting predictions of self focusing or defocusing. Using the R(22) line from a CO_2 laser to pump D_2O, the line centers of the lasing and absorbing transitions are separated by $\sim$ 300 MHz [9], in such a way that defocusing should occur. It would be possible to increase the efficiency of a D_2O amplifier by slightly focusing the R(22) pump beam, so as to compensate for the defocusing in the active medium. This is precisely what has been observed by Woskoboinikow et al.[2]

using an amplifier of up to 8 metres length.

5. CONCLUSIONS

We have shown that self focusing or defocusing of pump radiation in the medium of an optically-pumped FIR laser occurs, depending on the offset of the laser frequency from the absorption line centre. A general relationship between the absorption coefficient and self-focusing angle has been given. Consequently, for any FIR laser system, approximate focusing lengths may be calculated using readily-obtainable absorption data.

In the saturated regime, the magnitude of the angle is seen to be very insensitive to the beam intensity, and varies only with the square root of the frequency offset. This is important since (a) the frequency offset is not usually known with sufficient precision and (b) the laser line can be considerably wider than the absorption width. For a laser and absorption line in near coincidence it is likely that self focusing (on the high-frequency side) and self defocusing (on the low-frequency side) occur simultaneously. Characteristic self-focusing lengths are of the order of a few meters, and hence should be taken into account in the design of long axially-pumped FIR-laser amplifiers.

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FIGURE CAPTIONS

Figure 1 : Illustration of the derivation of equation (4). The caustic of a beam (between shaded lines) with axially-peaked intensity distribution (displayed on right) is shown in the vicinity of the beam waist.

Figure 2 : Normalised beam divergence and absorption coefficient versus frequency offset for different saturation parameters i.

Figure 3 : Experimental arrangement to measure self defocusing in CH_3F by means of the axial displacement of an air spark.

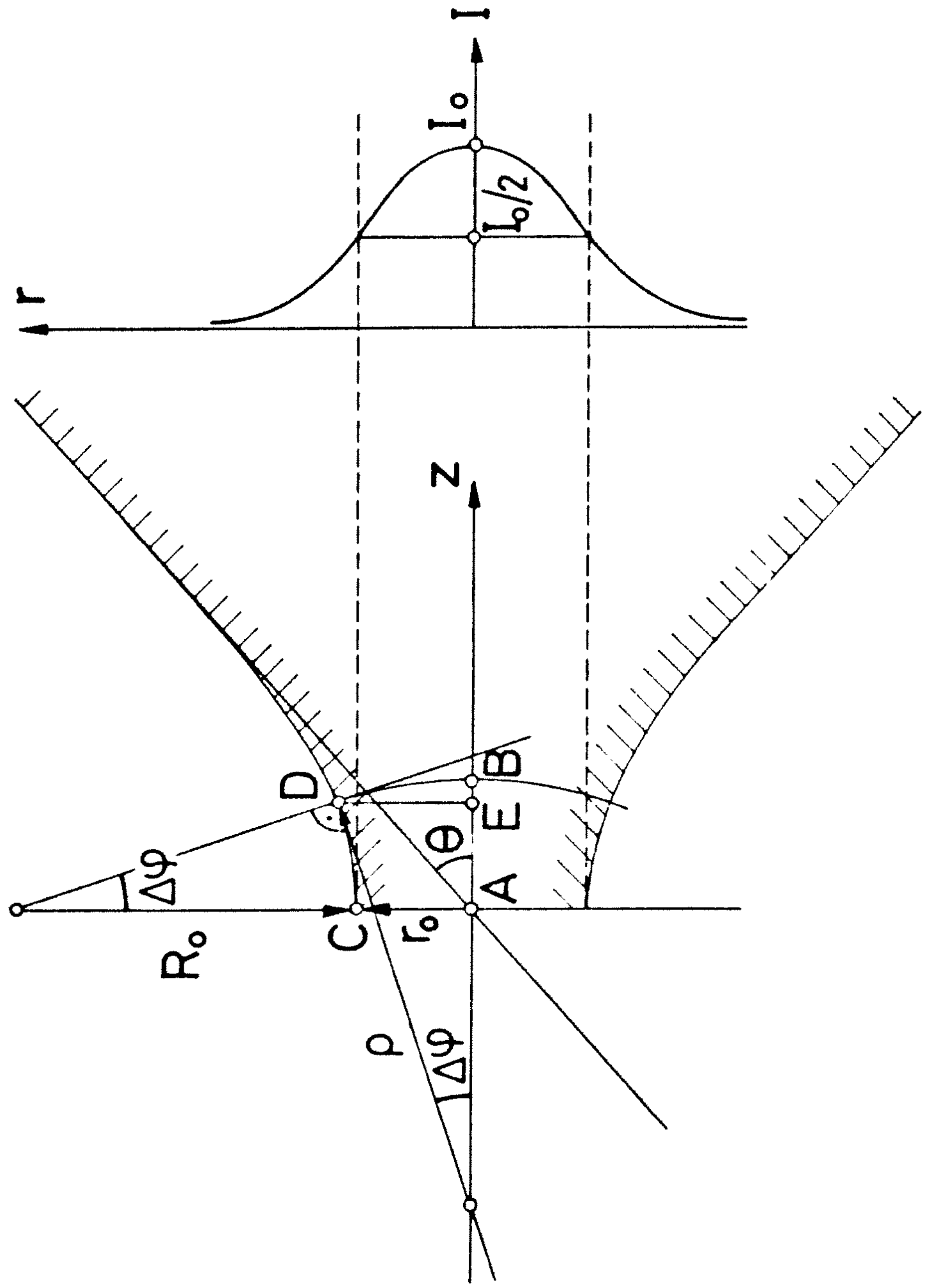
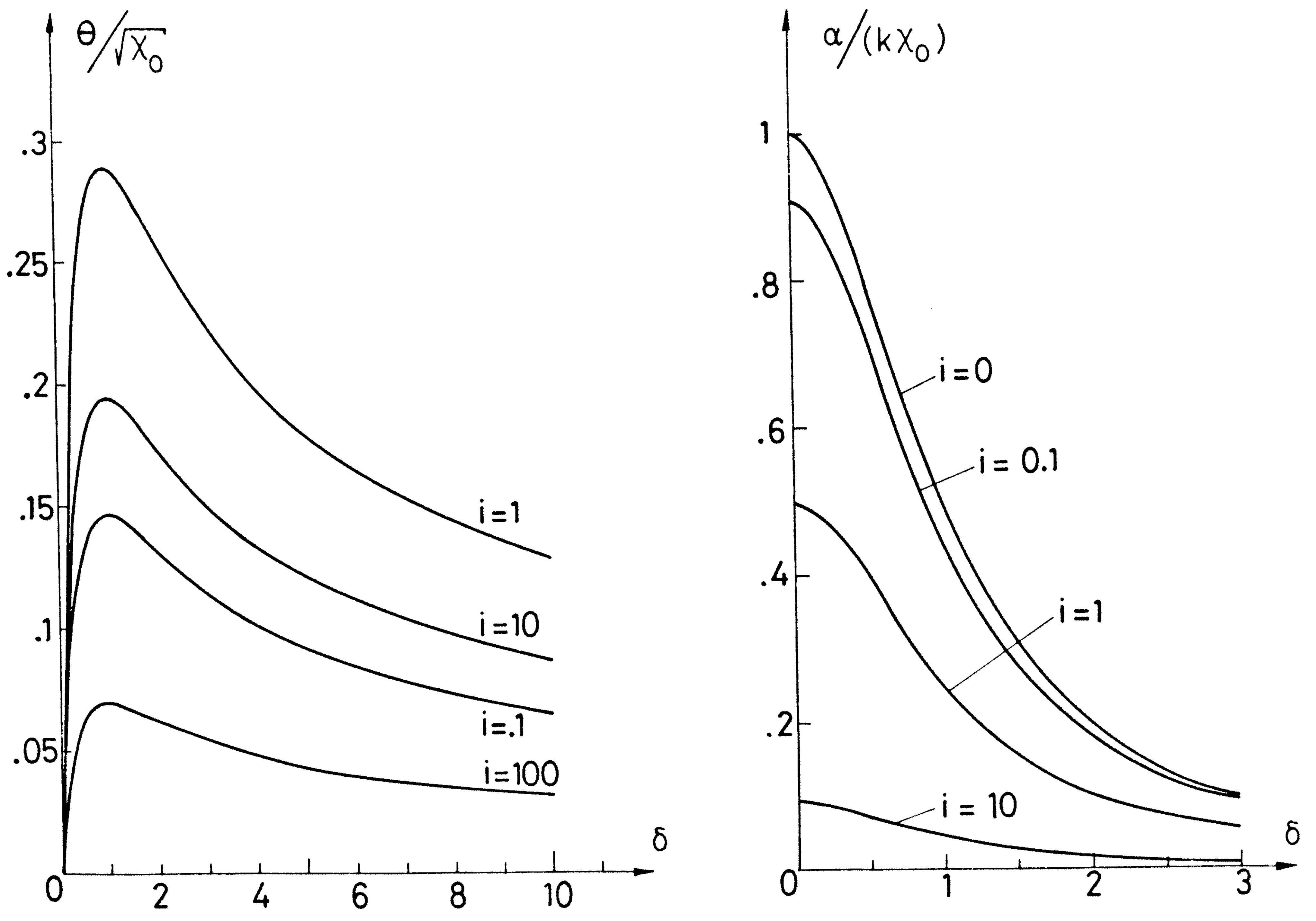


Fig 1



$\theta/\sqrt{\chi_0}$
.3
.25
.2
.15
.1
.05
0
2
4
6
8
10
δ
i=1
i=10
i=.1
i=100
$\alpha/(k\chi_0)$
1
.8
.6
.4
.2
0
1
2
3
δ
i=0
i = 0.1
i=1
i = 10

Fig 2

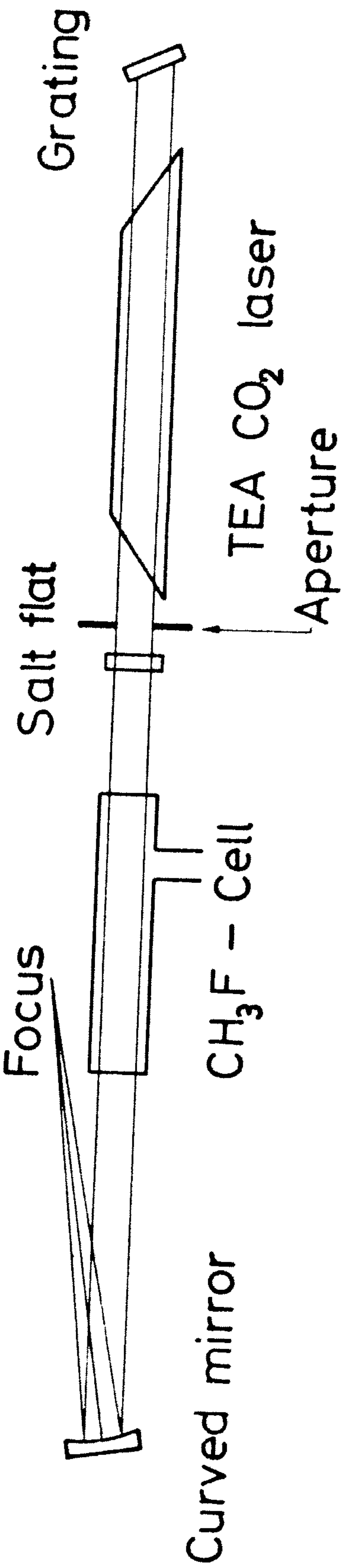

Grating
TEA CO_2 laser
Aperture
Salt flat
CH_3F – Cell
Focus
Curved mirror

Fig 3