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The Injection-Locked Single-Longitudinal-Mode TEA CO_2 Laser

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Abstract:

The development of a practically useful line-tunable TEA CO_2 laser giving reproducible smooth pulses from single longitudinal mode operation is described. Axial mode selection was achieved by injection of narrow band cw radiation into the laser cavity. Pulse energies of the order of 500 mJ were obtained for "stable" resonators and of 4 J for "unstable" resonators. Application to time-resolved experiments is illustrated.

I Introduction

The majority of TEA CO_2 lasers, used for example in infra-red laser photochemistry, operate on 5-15 longitudinal (axial) modes simultaneously and quite often on a number of transverse modes as well. Line selection by inclusion of a diffraction grating as one of the cavity reflectors does not change this situation since the resolution of the grating is insufficient to distinguish between axial modes which are typically separated by $\sim$ 60 MHz. The resultant output characteristics (1) include a 1-2 GHz spectral width, poor intensity definition due to a pulse time-development in the form of a burst of $\sim$ 1 ns pulses resulting from strong intermode beating (Fig. 1). These output characteristics are unsuitable for precise experiments requiring good definition of frequency and of intensity in both time and space. A desirable specification is 3-5 J, single mode, with a clean, reproducible pulse.

In a previous IPP report, Heckenberg (2) describes a number of methods of achieving single mode operation, in particular the hybrid low pressure-high pressure system. We summarise here some of the advantages and dis-advantages of various methods of solving the major problem which is that of selection of one longitudinal mode from several which can oscillate within the 2 GHz wide gain envelope of the standard TEA CO_2 laser (Fig. 2):

(i) Control of gain:

The Hybrid Low Pressure-High Pressure Laser.

References: (2), (3), (4), (5)

This method includes a low-pressure section, operating c.w., with a gain width $\sim$ 60 MHz, inside an extended resonator. By appropriately setting the c.w. gain level, the system can be made to oscillate on an SLM (single-longitudinal-mode). A number of such systems are in practical use. Disadvantage: the cw section is necessarily of $\sim$ 1 cm diameter and therefore poses optical adaption problems if combined with a large diameter TEA section. Thus, output energies are usually limited to $\sim$ 300 mJ, typical intensities being 100 KW - 1 MW.

(ii) Control of loss:

Spectral control of loss has been achieved through insertion of cells of SF_6 (6), or of Fabry-Perot interferometers (7) into the resonator and the use of three-mirror coupled resonators (8). Such systems can be adapted to large diameter gain sections but are sensitive in adjustment and not easily applicable to line-tuning.

(iii) Injection-locking:

This is the term we use for the selection of a SLM by injection into the laser resonator of a narrow band c.w. signal from a separate laser which causes one axial mode to compete preferentially with all other modes which must start from noise level. The advantages of the method are: (a) it uses well developed and engineered systems, (b) is non-critical in all parameters except frequency matching and in this respect not excessively demanding, (c) it is useable with grating line-tuning, (d) it is readily adapted to large diameter "unstable resonator" optics and (e) useable with some modification with existing PLF TEA laser systems. In complexity, it is comparable with a hybrid system; and like all single mode laser systems adequate control of resonator length is essential.

In the literature there are a number of reports on injection-locking, beginning with dye lasers (9) (10) and extending to CO_2 (11) (12) (13) (14). The latter group of papers contain apparently conflicting statements on the necessary conditions for mode selection - ranging from Izatt et al (12) who find "no requirement to match frequencies or stabilise cavity length" to the careful detail of both theory and experiment of Lachambre et al (13). Within the experimental limits set by different TEA laser arrangements, we relate to the conditions described by Izatt et al and develop the method into a practically useable system.

II Experimental

(a) Initial Experiments

For this purpose a standard 2-module Garching TEA laser with a non-dispersing 10 m radius concave mirror and plane coated germanium output couplers (R = 50% to 80%) were used. Unstabilised cavity lengths between 2 and 5 m were investigated.

The injection source was a line tunable Edinburgh Instruments PL3 CO/CO_2 laser system operating with CO_2 optics and a 17 Torr refillable charge of CO_2, N_2 and He. Maximum power output was $\sim$ 18 watts on P(20); more usually about 10 watts was employed. The mechanical stability of the laser (invar rod construction, etc.) gave long term frequency stability of the order of a few MHz over many minutes for single mode operation. This performance was checked with an Edinburgh Instruments scanning confocal interferometer. The laser output could be optimised or tuned over ∿ 20 MHz by piezo-ceramic control over the resonator length, nominally 1.70 m.

The PL3 output was injected into the TEA resonator (Fig.3) through the output coupler and arranged to sensibly fill the gain volume defined by a circular aperture. Clean (without temporal substructure) pulses representative of single mode operation were readily observed by photon drag detectors, e.g. Fig. 4, but found to occur inconsistently over time scales from seconds to minutes. This behaviour was later found to be caused by frequency drift of the TEA laser modes due to insufficient stability of cavity length. Nevertheless, the dependence of mode selection on aperture could be determined; it would found that best results were obtained with apertures ∿ 9-12 mm diameter; this defined essentially the TEM_{00} mode and sometimes one additional transverse mode. Partial locking could be observed up to 20 mm aperture diameter but the shape degraded. Mode selection was found to be not critically dependent upon either total cw power, (controlled with an Edinburgh Instruments LABCON attenuator over a dynamic range ∿ 1000:1) down to powers $\sim \frac{1}{100}$ watt, the injection beam optics or the output coupler reflectivity. Increasing the TEA resonator length to 4.25 mm gave a higher proportion of 'locked pulses' (up to 90%) but these remained rather unreproducible in shape and time delay. Locking was obtained when tuning the cw radiation from P20 to P22 and P24; this line tuning range might be infrared in the light of later knowledge. The alignment of the TEA laser has been mentioned e.g. (12) as critical; this was confirmed. We now believe this effect to be due to 'accidental' frequency matching.

(b) A grating-tuned semi-confocal TEM_{00} SLM injection-locked system.

Using the experience gathered from system (a) we developed the equipment to include a quartz-tube stabilised TEA resonator with the output coupler and grating rotation assembly mounted in marsine granite blocks. The output coupling coated germanium mirror was reduced to 25 mm diameter to fit an El Don piezo-translator which provided precise control of cavity length. Less precise control was also provided by mounting the grating on a translatable table which was used for some experiments. The grating was 75 lines/mm, concave, of radius 10 m, and obtained from Oriel. The apparatus, which included similar injection arrangements as in (a) through the output coupler, is shown in Fig. 5. The inclusion of two beam splitters enabled the output wavelength of both pulsed and cw lasers to be checked with Optics Engineering Spectrum analysers as well as providing probe and reference beams whose intensities were recorded on Rofin photon-drag detectors.

The most important experiments performed with this system concerned the translation of the output coupler in steps of $\sim$ 0.5 μm (50 V increments corresponding to $\sim$ 5MHz shift in the axial mode frequency). Simultaneously, the oscilloscope trace of the locked (or not-locked) pulse was photographed, Fig. 6, thus enabling the quality of the pulse, the delay time and the maximum intensity of the "gain switched spike" to be deduced. A plot of these quantities against piezo-voltage is given in Fig. 7. The "quality" of the pulse, i.e. its smoothness, is of course a subjective judgement - 'good' corresponds to the oscilloscope traces for 50, 100, 150 and 200 V for example. Thus a "mode selection range" of $\sim \pm$ 20 MHz about the nearest axial mode frequency is indicated. The pulse intensity and the delay can be precisely measured. A short delay always corresponds with a 'good' pulse. With frequency matching within ± 10 MHz of the axial mode frequency, the pulses are extremely reproducible and repeat better than 10 ns in time and to 1-2% in intensity. This is a most acceptable performance for use in precise measurement.

The pulse form shown in the 750 V-setting, Fig. 6, near the limit for injection locking was sometimes encountered. The time separation corresponds to a frequency separation of ∿ 7 MHz; this case may correspond to the selection of more than one transverse mode.

Output frequency could be line tuned by adjusting the gratings in both the TEA and the cw lasers. Essentially identical operation was obtained from R(10) to P(30).

We compare these results with the theory and experiment of Lachambre et al (13) in section III.

A summary of performance parameters is as follows:

Resonator length: 2.4 m, piezo-controlled.

Optics: Output coupler plane Ge, R = 65%, anti-reflected,
grating concave r = 10 m, 75 lines/mm R = 90%.

Aperture: Less than 15 mm, near output coupler, usually ∿ 12 mm.

Output energy: ∿ 200 mJ on P(20) with 75 nF charging capacitors (60 kV) (with 200 nF 500 mJ is possible). Approximately 5% of gain volume utilised.

Output power: ∿ 1 MW at peak on P(20).

Injection: stabilised single mode cw, 1-10 W.

Line Tuning: R(10) to P(30) at least.

Linewidth: Probably ≤ 20 MHz.

Reproducibility: Pulse to pulse ∿ 1-2% in intensity, < 10 ns in time.

(c) Large volume "unstable resonator" optics.

The arrangement described in (b), Fig. 5, while being useful and practical is inefficient in use of gain volume (∿ 5% used) and therefore limited in output energy to ∿ 500 mJ by the very nature and size of a TEM_{00} mode. A useful method for using a much larger proportion of the gain volume is to employ the so called "unstable resonator" configuration (15) (16). We were able to achieve injection-locked operation with relatively small modification to the system.

The same concave diffraction grating r = 10 m, diameter 43 mm, as in (b), was used, in combination with a convex mirror r = - 5 m, diameter 23 mm, combined in a near confocal resonator (L = 2.2 m). (Fig. 8). The cw radiation was injected by zero order reflection from the grating. Alignment of the cw beam was achieved by placing a small, thin, plane mirror on the grating surface and detecting the radiation at the convex mirror with a uv image plate. No more precise adjustment was necessary.

The results of injection are shown in Fig. 9. The upper traces show some residual mode beating and the lower the results of adjusting the length of the cavity piezo-electrically, when good quality clean pulses were obtained. In the second low trace the locking persists for ∿ 1 μ sec before beating sets in. Energy measurements indicated ∿ 4 J per pulse. No aperture was required the better directional quality of the "unstable resonator" apparently being sufficient to limit the number of transverse modes - (although mode beating between two modes was also observable on occasion).

The concave grating could be rotated to give line-tuning; its alignment was rather critical but proved to be practically useable. The geometry of injection is of course subject to re-alignment for

for each new grating setting and therefore not optimum. Other injection schemes, e.g. using a semi-reflecting convex mirror or through a small aperture in this mirror, should provide better solutions.

The output beam quality and shape is shown by the mode pattern depicted in Fig. 10. Initially an annulus with divergence of 5 m rad is observed. Focussing with a 10 m concave mirror concentrated $\sim$ 50% of the energy in a 0.5 cm diameter circle at $\sim$ 18 m distance giving an intensity > 50 MW/cm^2.

III Theory and Interpretation

There are three interesting physical questions concerning mode selection by injection locking:

(i) Under what conditions can one mode be induced to grow at the expense of all others?

(ii) What must be the geometrical relation between the injected beam and the mode characteristics of the growing wave?

(iii) What is the frequency relation between the injected beam, the TEA laser mode frequencies and the frequency of the final output?

Early papers, for example Vrehen & Breimer (9), describe the use of injection-locking with dye lasers as a method for mode selection. Several papers

(11) (12) (13) (14) describe the extension to the case of CO_2 and give theories of the mechanism. In some cases of this early work the theory is insubstantiated, or difficult to assess. A convincing treatment is given by Lachambre et al (13) on which we base the present interpretation. For typical TEA CO_2 laser operating conditions, the weak spontaneous noise, E_s^n, emitted at cavity frequencies has a value of the order of 4×10^{-12} W/cm^2. The theory describes the transient effects resulting from the competition between such modes and the strong (mW/cm^2 - W/cm^2) injected radiation, E_i, at the "master oscillator" frequency. With an injected signal, the total cavity field is the coherent sum of all the amplitudes E_s^n and E_i. Only the low frequency envelope affects the population inversion, thus the total photon density is calculated from an incoherent superposition of the various cavity amplitudes and used with a conventional 4-level model of the CO_2-N_2-He system to calculate the true dependence of the populations and hence predict pulse shape and time-dependence of inversion. The calculations are made as a function of frequency difference between E_i and E_s^n at the nearest axial mode frequency.

Qualitatively, during gain build-up, both the noise signal at the mode frequency ω_m and the injection signal at the master oscillator frequency ω_i circulate simultaneously in the cavity. Competition sets in between the two sets of waves as to which will grow more rapidly and depopulate the gain medium before the other enters the saturation regime. The injection field is much larger than the spontaneous emission but experiences a much smaller gain because it, in general, suffers a phase change on each cavity traversal (due to a finite $\omega_i - \omega_m$) and thus no longer adds in phase with E_i.

Thus, depending on the actual conditions, injection level and frequency difference $\omega_i - \omega_m$, the latter being more critical, either can reach saturation first. The time varying phase of the growing wave adds to the fixed phase injection signal amplitude vectorially to produce a resultant changing in amplitude and phase with time. The time-varying phase is equivalent to a frequency modulation that pulls the field frequency towards the nearest axial mode. The amount of pulling varies with time and depends on instantaneous gain, detuning frequency and injection level. Lachambre et al calculate that for a typical mode-selection situation, the frequency pulling takes place during the first 0.5 μ sec and is essentially complete by the time the laser pulse begins, the frequency thereafter remaining at the mode frequency. The most measurable parameter predicted is the "mode selection range". This is not sensitive to injection level - changing by $\sim$ 20% over two orders of magnitude of cw intensity. Maximum selection range is limited to about 40% of the inter mode frequency spacing for practical injection intensities. Our experimental finding of $\pm$ 20 MHz in 60 MHz mode separation for adequate mode selection is in good order of magnitude agreement with Lachambre et al in both theory and experiment.

We can comment on the paper of Izatt and Mathieu and Budhiraja (12), and on other work with unstabilised cavities, such as our section (a):- if the cavities are long so that the axial mode spacing $\Delta\nu = c/2L$ is small - the cw laser can give two axial modes - frequency mismatch can then be relatively small and give a reasonable probability of mode selection. Such systems will have limited reproducibility, however, if the length is unstabilised.

Rather limited information is available either theoretically or experimentally concerning the requirements on geometrical form of the injection beam. Critical wavefront matching is unnecessary according to experimental result but since small power densities easily compete with spontaneous noise small amounts of scattered radiation may play a role.

IV Conclusions

We have shown that longitudinal mode selection by injection locking is a practical and useful technique for improving the precision of measurement possible with grating-tuned TEA CO_2 lasers. The performance is best illustrated by oscilloscope photographs of absorption measurements on SF_6. Absorption could be measured to a few percent with a time resolution of < 1 ns, from just one pulse - Fig. 10. (Further details are given in (17)). Pulse reproducibility was excellent. The major experimental precaution necessary for such operation is the stabilisation and adjustment of the TEA CO_2 laser cavity length. Otherwise all parameters were found to be non-critical (although the injected frequency must also be stable between pulses). Mode selection was obtained satisfactorily within ± 20 MHz of an axial mode frequency - about 30% of the mode separation in frequency. Using conventional 'stable' resonator optics, the gain volume that could be utilised was limited by the necessity of selecting a near TEM_{00} mode by inclusion of an aperture. As a result only 5-10% of the available volume from the standard Garching PLF TEA CO_2 module could be used with an energy limitation of $\sim$ 500 mJ. In terms of fluence, however, the output $\sim 2J/cm^2$ begins to approach damage levels for optical components.

In a short investigation, the extension to multi-Joule energies was explored using "unstable" resonator optics. Successful mode selection by injection was achieved and useable clean pulses observed with pulse energies not less than 4 J, the system being line-tunable by means of a concave diffraction grating. It should be noted that the grating alignment, in particular, was very critical in this case and further studies of alignment and optical tolerances (15), better adjustment facility, optimisation of grating and cavity parameters and injection geometry are desirable. Nevertheless, with a modest development programme, routine availability of 4-5 J SLM pulses seems entirely possible.

Both the 'stable' and 'unstable' resonator configuration used have the full line-tuning capability of the cw and TEA CO_2 laser.

Injection can also be applied to mode-locking of pulse trains using switched nanosecond cw sources (18).

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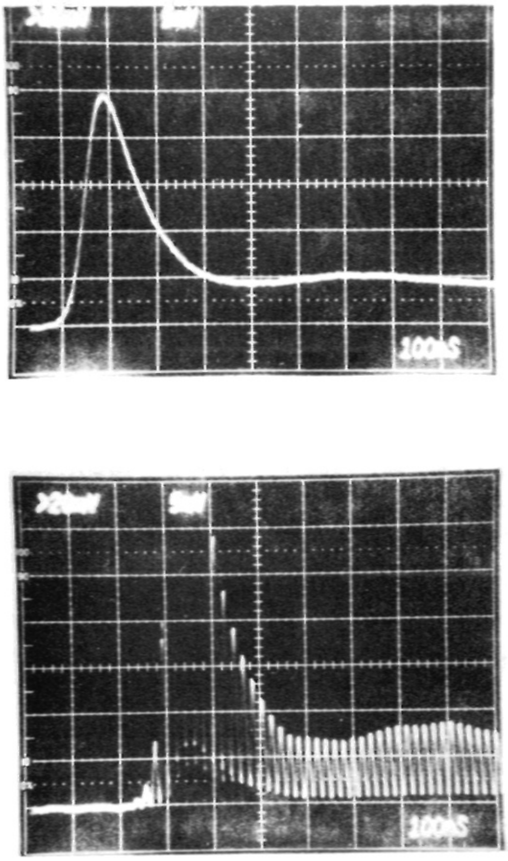

Fig. 1 Mode beating in a multimode TEA CO_2 laser compared with SLM pulse.

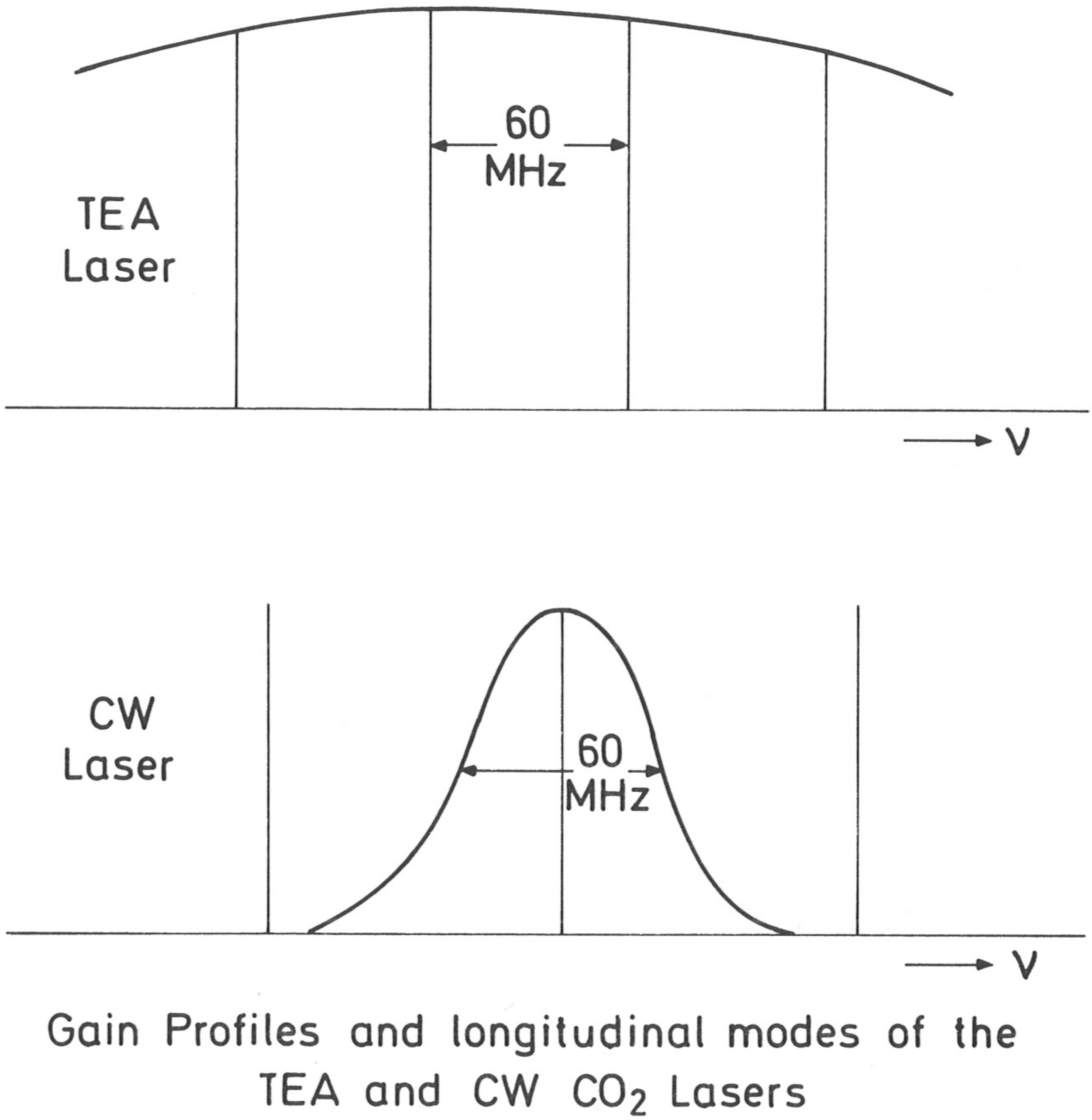

Fig. 2 Gain linewidth in TEA and cw CO_2 lasers.

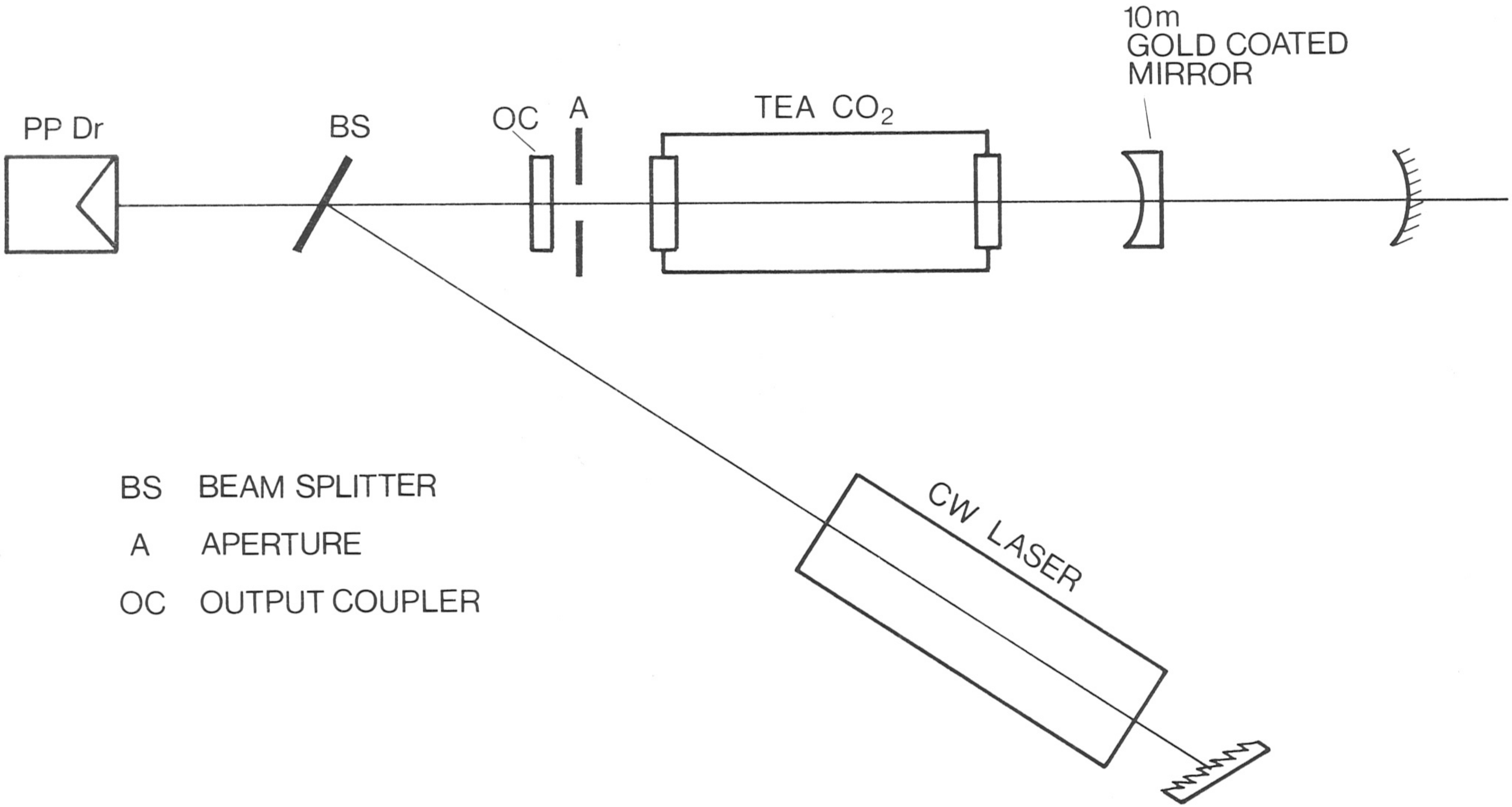

Fig. 3 Experimental arrangement for injection locking with non-dispersive cavity reflectors.

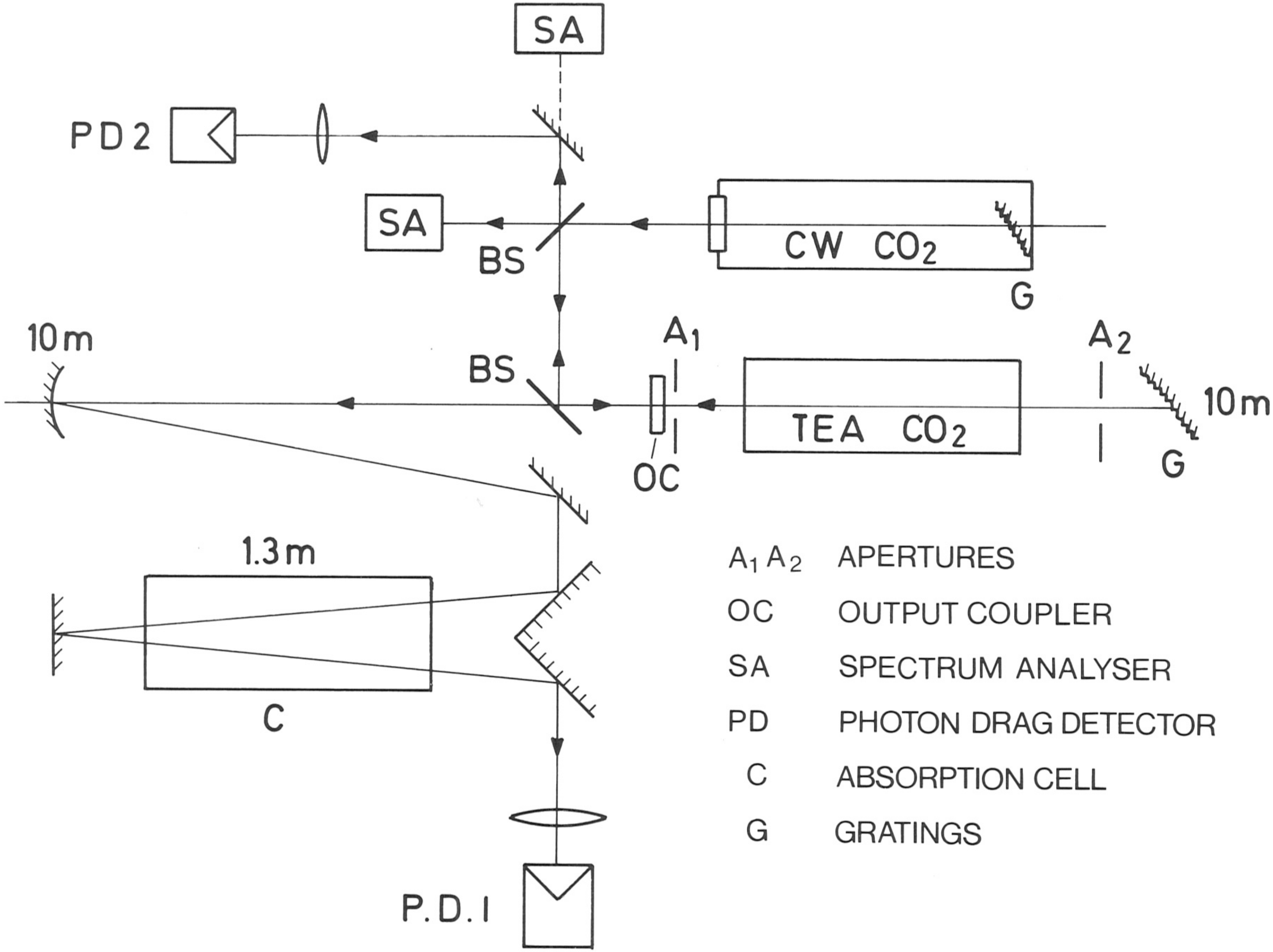

Fig. 4 Grating-tunable injection-locked single mode CO_2 laser system with arrangements for absorption measurements.

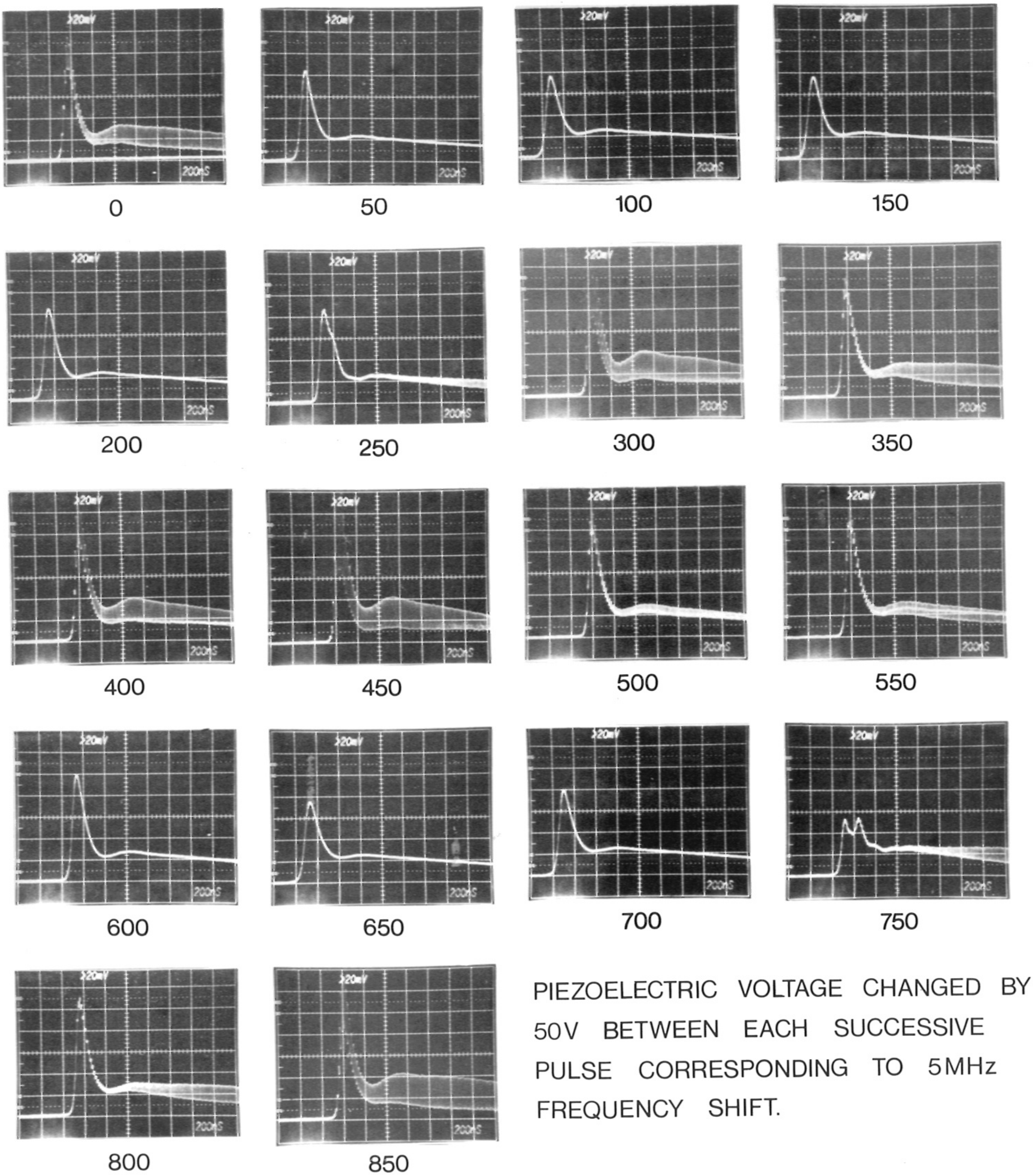

Fig. 5 Pulse form variation as a function of piezo-translator voltage.

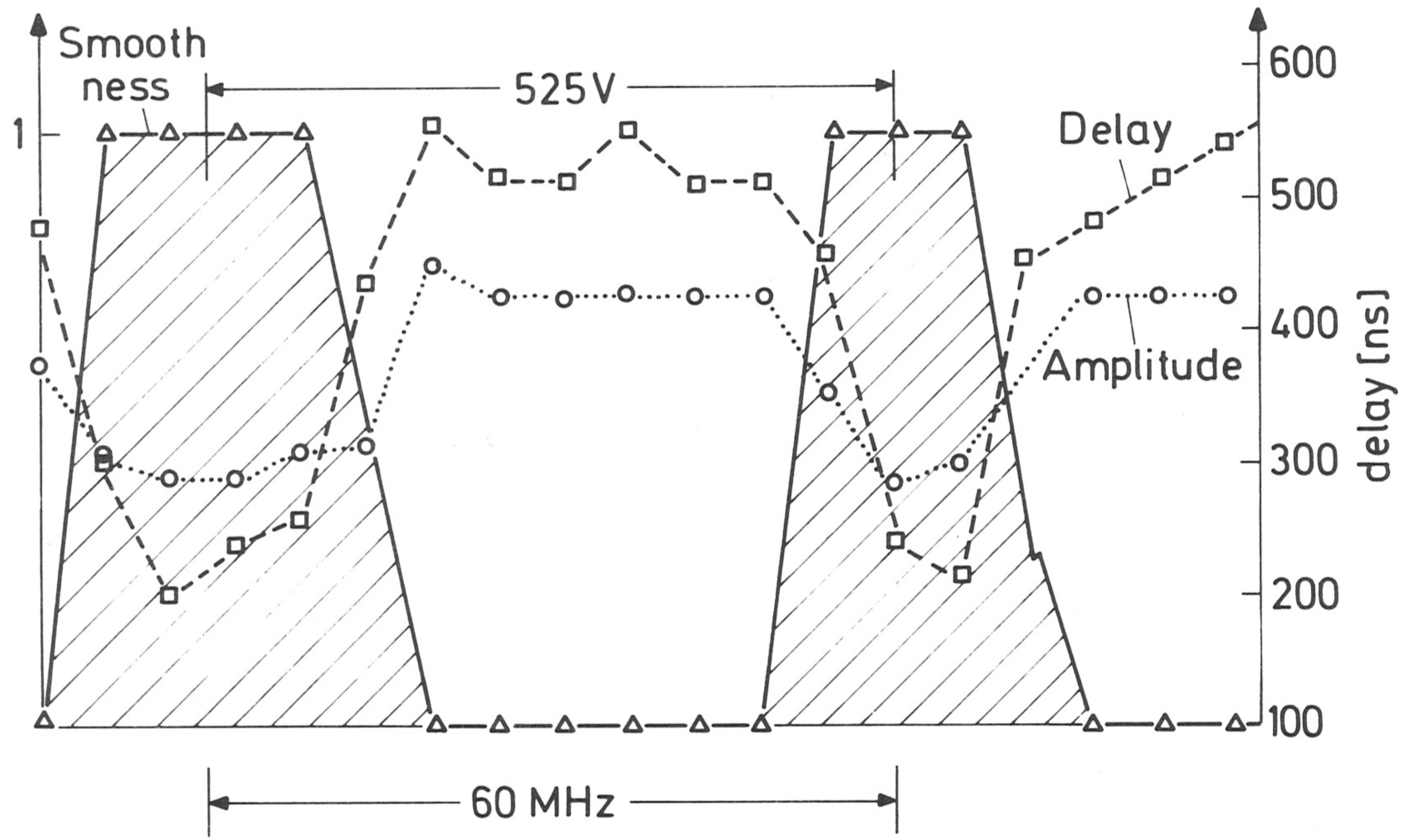

Fig. 6 Mode selection ranges, pulse time delay and peak intensity as function of detuning frequency.

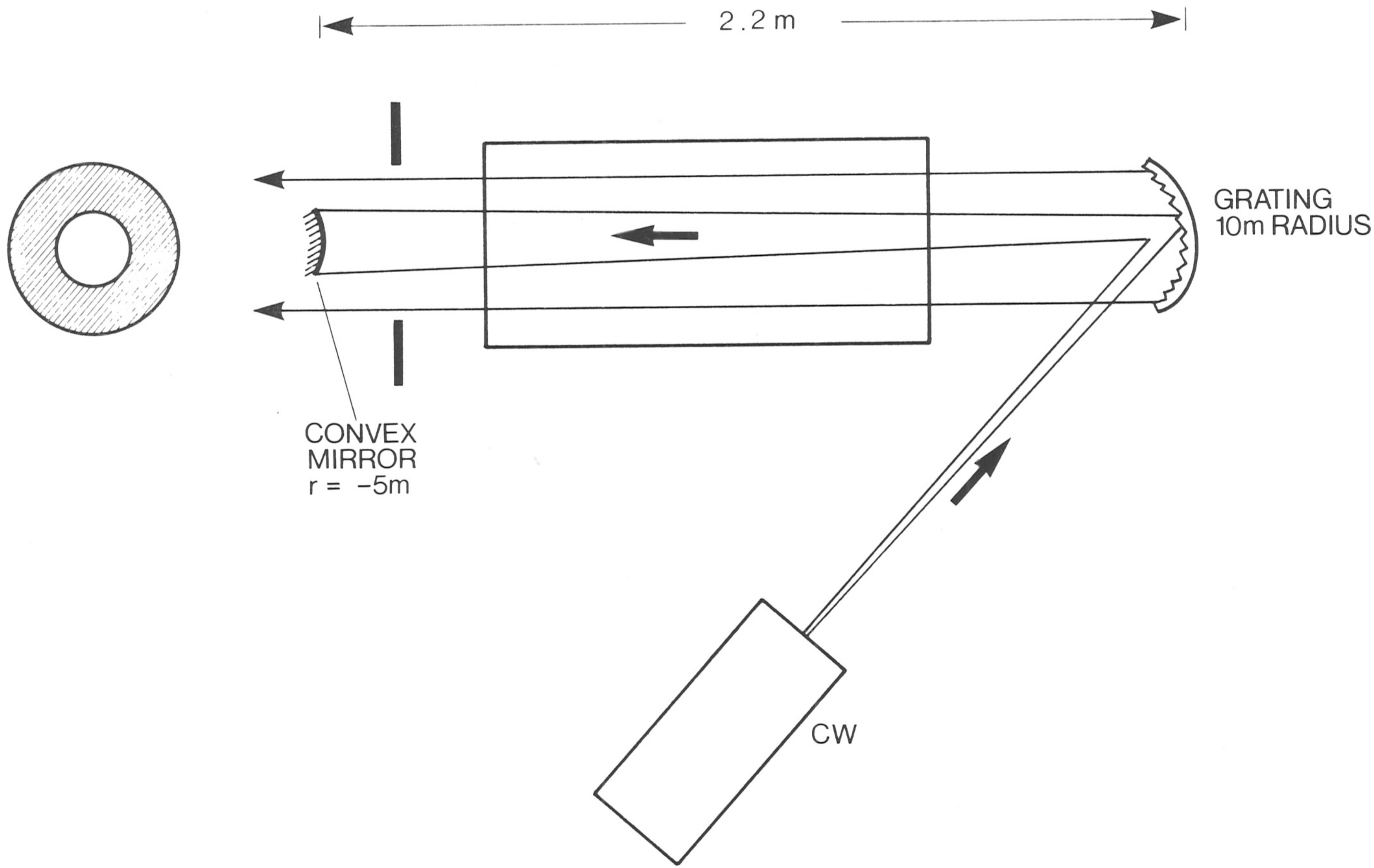

Fig. 7 Injection into resonator with "unstable" confocal optics.

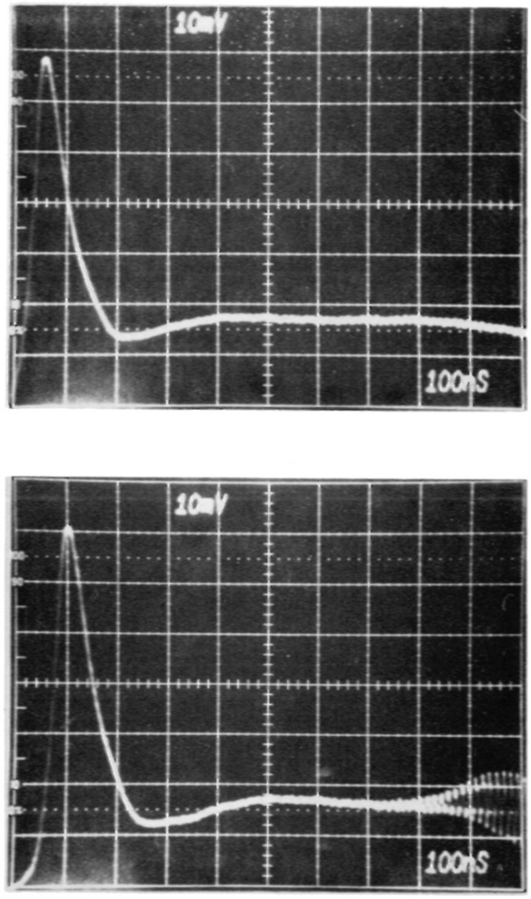

Fig. 8 Smooth pulses from unstable resonator.

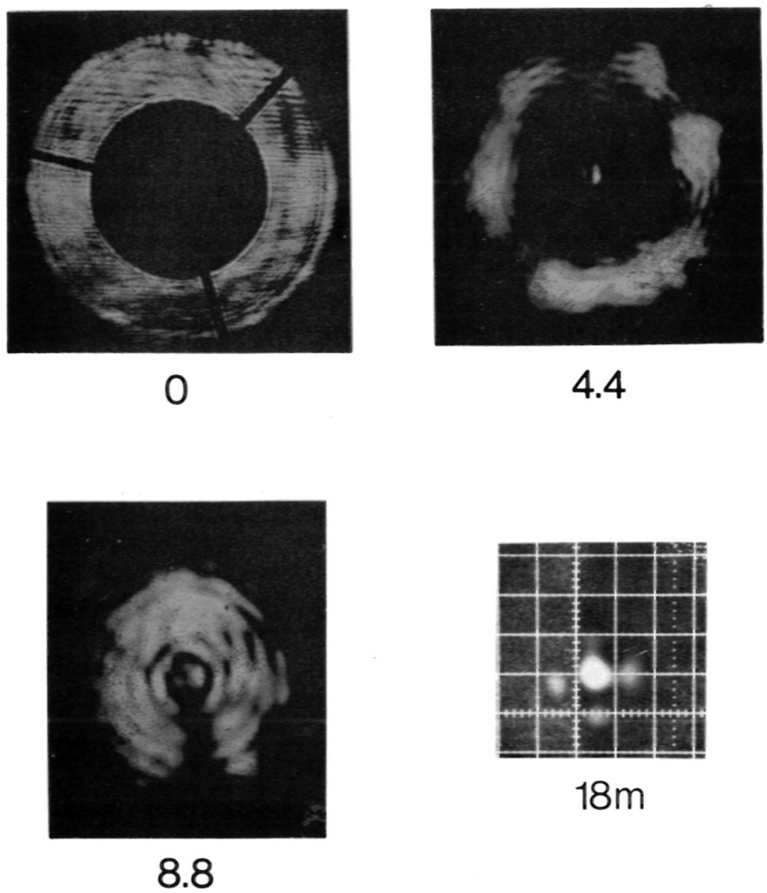

Fig. 9 Output beam form from unstable resonator.

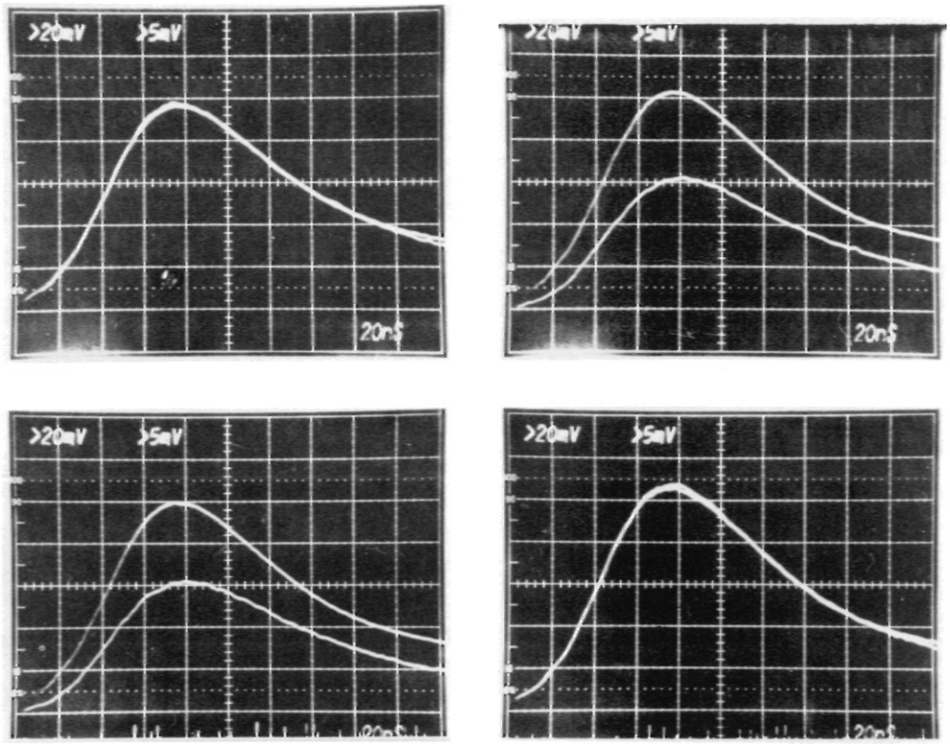

Fig. 10 Pulse form for SF_6 absorption measurements.