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DESIGN PARAMETERS FOR AN OPTICALLY-PUMPED FIR - LASER OSCILLATOR

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ABSTRACT

Gain and loss coefficients based on a coupled three-level system, including single and double photon transitions, are applied to study an optically-pumped FIR laser oscillator. Guidelines for optimising such an oscillator with respect to any one of several interesting parameters (peak-power, energy, efficiency) are given. An important result is that too much pump power results in splitting of the gain line and thus to a decrease of coupling into the fixed-frequency modes of the oscillator.

1. Introduction

Optically pumping a far infrared (FIR) laser is a complex process. Without a detailed understanding of the pumping mechanism it is not possible to optimise the design of a FIR laser oscillator other than by experimental trial and error. The theory of optical pumping in terms of rate equations has recently been developed and it is the purpose of this paper to point out some easily applicable general relationships which allow the experimenter to choose an optimum resonator and pump pulse for his FIR laser system.

Rate equations for an optically-pumped FIR laser have been developed by several groups[1,2,3]. Take for example the model of Temkin and Cohn[1] which describes a CH_3F laser. In this model the rate of change of five population densities are investigated under the influence of pump and FIR beams and collisional processes. These are the population densities of the three energy levels associated with the pump and FIR transition and the sum of the molecules in the rotational levels of the ground and excited vibrational levels, respectively. An equilibrium solution yields a saturable pump absorption coefficient and a FIR gain which is saturable with respect to both the FIR and the pump intensities. Most efficient operation is obtained with both a saturating FIR beam intensity and a saturating pump.

Lineshapes of optical transitions are either Gaussian or Lorentzian, depending on whether the line is Doppler or pressure broadened. When

FIR systems that operated with large offsets between pump frequency and absorption line frequency were discovered, this was explained by off-resonant pumping, otherwise known as wing absorption[4]. It was first pointed out by Chang and McGee[5] that a Raman-type two-photon process might be responsible for the observed behaviour. This idea has since been backed up by experimental evidence[6]. It is evident from the treatment of Panock and Temkin[2], which we intend to follow here, that laser and Raman-type processes are not two completely different uncorrelated effects. Theoretically, both follow from a quantum-mechanical treatment of a coupled three level laser system, first presented by Javan[7]. The FIR gain of a three level system has two components : a laser and a Raman component. The latter is not negligible even for the case of resonant pumping. Also included in the model are two power-dependent effects: splitting of the gain profile due to the ac-Stark effect (Rabi splitting) at high pump powers and line broadening caused by intense FIR radiation. These effects have important consequences. With the simple rate equation model one finds a FIR gain which is almost independent of pump intensity for a saturating pump, whereas there is actually an optimum pump power for maximum FIR gain. A further increase in pump intensity is not only unnecessary but is, in fact, disadvantageous because the gain at a fixed frequency is decreased and can become insignificant. This is a manifestation of the ac Stark effect splitting up the line into two distinct peaks. If the FIR frequency could change to follow one of the peaks as it moves away spectrally from the low intensity line center, the gain would remain high. Fortunately this problem is alleviated at higher FIR intensities by power broadening which smears out the gain profile over a large spectral range. We will demonstrate later that a well-defined balance between

Rabi splitting and power broadening, and hence between pump and FIR intensities, has to be found in order to maximise the gain.

2. FIR Gain as a Function of Pump and FIR Intensities

The FIR gain coefficient g_f and the pump beam absorption coefficient g_p for the inverted vee configuration in Fig. 1a. can be expressed in the following way[3]

$$\begin{aligned} g_f &= \sigma_{32} \left[(n_3-n_2)\, F_1 + (n_1-n_2)\, F_2\right] \\ g_p &= \sigma_{31} \left[(n_1-n_3)\, f_1 + (n_1-n_2)\, f_2\right] \end{aligned} \qquad (1)$$

where σ_{ij} is the homogeneous cross section at line center and $F_{1,2}$, $f_{1,2}$ are complicated functions of pump and FIR intensities and detunings from line center. Each expression contains two terms : a term proportional to the population difference of the two levels involved in the transition ($n_3 - n_2$ for the FIR and $n_1 - n_3$ for the pump), describing the laser or single photon process, and a term proportional to the population difference of the initial and final level of a two-photon transition. In their discussion of these equations, Panock and Temkin[2] analysed in detail the lineshape of the gain coefficient for resonant and off-resonant pumping. They find for given beam intensities that the gain peak for off-resonant pumping can exceed the maximum gain for resonant pumping. The position of the gain peak in the frequency spectrum, however, varies with pump and FIR beam intensities due to power broadening and Rabi splitting. Hence a mode in a resonator with a fixed frequency, optimised for a given set of intensities, will not always see an optimum gain during its build-up time. In fact our own calculations indicate that optimum performance of a resonator is always obtained with resonant pumping. It has been shown experimentally[8] that the most commonly used pump source, the CO_2 laser, can easily be shifted in frequency by up to ~2GHz to bring it into coincidence with absorption lines of FIR lasing materials. Consequently, this paper deals mainly with resonant pumping.

An equilibrium solution of the rate equations is possible if the collisional relaxation rates of all three energy levels involved are considerably faster than the characteristic time scales for changes in the beam intensitiy. This is, for example, the case for a D_2O laser. The equations are complicated in the general case, but reduce to a simple form for resonant pumping. On line center one obtains for the gain coefficients

$$g_f = 9g_f^{ss} \frac{P_p}{(1+P_p+P_f)(1+4P_p+4P_f)}$$

$$g_p = g_p^{ss} \frac{1+P_p+4P_f}{(1+P_p+P_f)(1+4P_p+4P_f)} \qquad (2)$$

We have normalized the beam intensity of the pump (P_p) and of the FIR (P_f) with respect to the corresponding saturation intensities $I_{sat} = \frac{1}{4\pi} \frac{ch^2}{2\mu_{ij}^2} \Delta\nu_H^2$, where μ_{ij} is the transition dipole moment and $\Delta\nu_H$ the homogeneous linewidth. We have introduced the small signal absorption coefficient g_p^{ss} and the small signal ($P_f \ll 1$) FIR gain coefficient g_f^{ss} for optimum pumping ($P_p = \frac{1}{2}$).

For resonant pumping the FIR power extractable per unit length is shown in Fig. 1a as a function of pump power for several different FIR intensities. For any fixed FIR power there is clearly a peak for a certain pump power. A higher pump power is not only unnecessary, but detrimental to efficient operation. The position of the peak shifts towards higher pump intensities for increasing FIR powers. In the highly saturated regime ($P_p \gg 1$, $P_f \gg 1$) we obtain from eq. (2)

$$g_f \approx 2.25 g_f^{ss} \frac{P_p}{(P_p+P_f)^2} \qquad (3)$$

from which it can easily be deduced that the gain for a given FIR power is a maximum for $P_p = P_f$.

It is interesting to compare the contributions of single and double photon transitions to the gain. These are shown in Fig. 1b) for the same conditions as in Fig. 1a). It is somewhat surprising to find that the double photon component behaves in a way that would be expected from the single photon component whereas the main effect of the single photon component seems to be to counteract the Raman gain. At low FIR intensities the Raman gain increases monotonically with pump intensity until it eventually saturates. The laser gain, at first, increases too before reaching a maximum and then, falling below zero, asymptotically approaches a negative value of gain of the same magnitude as the saturated Raman gain. Thus it is the single photon transition which is responsible for the decrease in the total gain at high pump powers. For the conditions where the laser gain is negative one would espect that there is no longer a population inversion between levels 2 and 3. This is, however, not true. The apparent contradiction is resolved in considering the whole spectral gain profile instead of its center-line value only. The total integrated laser gain is positive due to off-resonant peaks even for a negative center-line value, as long as a population inversion exists.

At high FIR powers the conditions are more complex. With reference to Figure 1b the laser gain on line center is now practically always negative and only due to the Raman gain is a positive total gain still possible.

We have also investigated the line profiles of the two FIR gain components (laser and Raman) separately for on and off resonant pumping. It is not straightforward to give a physical interpretation of these results, hence we will only list a few observations. If either the pump power or the FIR power or both are increased, in the case of resonant pumping, the single photon component of the gain splits into two peaks with a trough in between,which becomes negative even for moderate power densities. The Raman component also splits with high pump powers, but far less readily and the center line gain remains almost always positive. Also the Raman gain is hardly affected by the FIR intensity and shows no splitting at high FIR fields. The situation where the total gain consists of roughly equal single and double photon components is very rare. Normally the total gain is due to either Raman or laser process with the other one contributing very little or in fact counteracting it.

This behaviour is clearly visible with off-resonant pumping where laser and Raman gain peaks are well separated. For low pump and negligible FIR intensity the Raman gain has a peak which is offset from the center line frequency by an amount corresponding to the offset of the pump frequency from the frequency of the absorbing transition. The laser gain has a peak on line center, where the Raman gain is negative, counteracting about 50% of the laser gain. If either or both intensities are increased, the laser gain also develops a negative peak at the position of the Raman line. Despite these negative peaks the total gain is always positive. It seems that the balance between absorption and stimulated emission according to the population distribution is not maintained for laser and Raman gain separately, but only for the sum of both processes. Whenever stimulated emission due to one of the mechanisms is favoured, this emission grows even further at the expense of the other. Laser and Raman gain are obviously closely intertwined and cannot be easily separated either in theoretical treatments or in experiments.

With these observations the behaviour on line center for resonant pumping, shown in Fig. 1b is now understandable. At low intensities single and double photon processes both contribute to the gain. At higher powers the levels split due to the ac Stark effect. The emission of a FIR photon due to the laser process is decoupeld from the absorption of pump photons and hence its frequency will readily follow the splitting. The Raman process, however, depends on the pump frequency, which is fixed, and hence, for resonant pumping, will not readily move off line center. The Raman effect will thus provide the gain on line center and and will force the laser contribution to become negative, according to the observations made above.

3. Buildup of FIR Radiation in an Oscillator

The buildup of FIR power in an oscillator can now be studied by simultaneously solving the differential equations for the beam propagation of pump and FIR beams within the resonator. Neglecting the effects of spatial mode structure and of the standing wave pattern formed by the forward (P_f) and backward (P_b) propagating FIR beams, we obtain

$$\frac{dP_\ell(x,t)}{dx} = -g_\ell(P_\ell,P_f,P_b)\, P_\ell(x,t)$$

$$\frac{dP_f(x,t)}{dx} = g_f(P_\ell,P_f,P_b)\, P_f(x,t) + \gamma n_3(x,t) \tag{4}$$

$$-\frac{dP_b(x,t)}{dx} = g_f(P_\ell,P_f,P_b)\, P_b(x,t) + \gamma n_3(x,t)$$

We have assumed that the resonator mirrors do not reflect the pump beam and hence there is no backward propagating pump wave. The three equations are coupled via the gain coefficients and the FIR beams are also connected by the boundary conditions

$$P_f(0,t) = R_1 \; P_b(0,t)$$
$$P_b(\ell,t) = R_2 \; P_f(\ell,t) \qquad (5)$$

where R1, R2 are the mirror reflectivities and ℓ is the resonator length. In the set of equations (4) we have also introduced a term responsible for spontaneous emission, proportional to the population density n_3 of the upper level of the FIR transition. The proportionality factor γ depends on the geometry of the resonator.

We have numerically solved these equations with a simple explicit difference scheme. The gain coefficients vary sufficiently slowly as functions of the beam intensities to guarantee stability of the numerical scheme. A detailed account of the parametric study of FIR oscillators performed with this code would exceed the scope of this paper. Hence we will restrict ourselves to a few general remarks, valid for parameters which have been varied around the following average values : resonator length 1 m, Gaussian pump pulse of 50 ns width. The results are : (1) Optimum performance is obtained with resonant pumping. (2) If the small signal gain g_f^{ss} does not exceed a certain limit ($g_f^{ss} \ell \approx 10$), the output pulse in effect consists of amplified spontaneous emission and depends strongly on the geometric factor γ. (3) A periodic structure is usually superimposed on the output signal with a period representing the roundtrip time in the resonator. The amplitude of the superimposed signal grows with decreasing mirror reflectivity. It can be shown that this periodicity is not an artificial numerical feature. Due to the limited number of available roundtrip times in the resonator ($\sim$10), discontinuities of the intensity profile such as due to the output coupling cannot be smoothed out. (4) The dependence of output power on output coupling is weak. A mirror reflectivity of $\approx$ 50% is optimal. (5) The output power is proportional to the small signal gain. Maximum output power is obtained for pump intensities of about 10 times saturation intensity. If the pump power is further increased, the FIR pulse splits up in time into a double pulse with smaller amplitude and reduced total energy.

4. Optimization of a Resonantly-Pumped FIR Oscillator

We will now introduce some simplifications and try to develop a somewhat more intuitive picture of resonant pumping. We first restrict ourselves to resonator lengths which are considerably shorter than the spatial extent of the pump pulse (e.g. a 100 ns pump pulse and a 50 cm long resonator), so that the intensity of the pump beam and the sum of the intensities of forward and backward travelling FIR beams do not vary considerably within the resonator and for one roundtrip. In this case the temporal change of FIR intensity in the resonator can be written as

$$\frac{dP_f}{dt} = c\ P_f\ (g_f(P_p,P_f) - \alpha) \tag{6}$$

with a loss α. The loss is mainly due to output coupling and depends on the mirror reflectivities in the following way[9]

$$\alpha = - \frac{\ln(R_1\ R_2)}{2\ell} \tag{7}$$

The power density P_f in Eq. 6 is the sum of contributions from forward and backward propagating waves. In order to obtain the intensity outside the resonator, the intensity of the forward propagating wave $P_f/(1+R)$ has to be multiplied by the transmission factor $1-R$, where R is the reflectivity of the output coupler.

We first consider a lossless resonator and study its performance in the highly saturated regime. The gain is then given by Eq. (3) and we obtain from (6)

$$\frac{dP_f}{dt} = 2.25 c g_f^{ss} \quad \frac{P_p\ P_f}{(P_p+P_f)^2} \tag{8}$$

As already mentioned, the gain in this case is a maximum for $P_p = P_f$.

Inserting this into (8) we obtain

$$P_f = P_p = {}^9/_{16}\, c g_f^{ss}\, (t - t_o) \tag{9}$$

If a pump pulse in the form of a linear ramp according to (9) is applied to a short, lossless resonator, the FIR intensity grows linearly to any desired value.

The assumption of a linearly-rising pump pulse is not unreasonable, but its slope, i.e. rate of change of intensity, is not easily adjustable and may not correspond to the value given by (9). Hence we consider solutions of (8) with linear ramp-type pump pulses $P_p = \beta(t - t_o)$ with non-ideal slope β. The FIR pulse is then also found to be a linear ramp with slope

$$\gamma = {}^3/_2 \sqrt{c g_f^{ss} \beta} - \beta \tag{10}$$

Fig. 2 shows the slope of the FIR pulse versus the slope of the pump pulse. It is found for pump pulses with steepening ramps that the slope of the FIR pulse increases to a maximum at $\gamma = \beta = 9/16\ c\ g_f^{ss}$ (Eq. 9) and then decreases to zero at $\beta = 9/4\ c\ g_f^{ss}$. Given a linearly rising pump pulse of fixed duration, Fig. 2 tells us which pump power to choose to obtain maximum FIR power. We find again that too much power is not only unnecessary, but in fact undesirable.

Usually, the energy and power in the extracted pulse are of prime interest and not the intracavity values. Hence we will re-examine Eq. (6) using the gain value given by (3) and the cavity loss by (7). Knowing already that optimised conditions are obtained for $P_\ell = P_f$, we first solve the equation for this case and obtain

$$P_f = 9/16 \; g_f^{ss}/\alpha \; \frac{1-R_1}{1+R_1} (1-e^{-\alpha ct})$$

$$= 9/8 \; g_f^{ss} \ell \frac{R_1-1}{\ln R_1 R_2} \; (1-(R_1 R_2)^{t/\tau})/(1+R_1) \qquad (11)$$

where τ is the cavity roundtrip time $2\ell/c$. The factor $\frac{1-R_1}{1+R_1}$ relates the intensity coupled out, through the mirror with reflectivity R_1, to the intracavity value. Mirror R_2 is highly reflecting. According to equation (11) the FIR beam intensity first grows linearly with slope $\frac{dP_f}{dt}(0) = \frac{9}{16} c \; g_f^{ss} \; (\frac{1-R_1}{1+R_1})$ and then approaches asymptotically a maximum value of $9/8 \; g_f^{ss} \; \ell \frac{R_1-1}{\ln R_1 R_2} \; \frac{1}{1+R_1}$. For a given R_2, R_1 can be chosen to maximize the expression $(\frac{R_1-1}{1+R_1})/\ln R_1 R_2$ to obtain maximum FIR power. This maximum is, however, rather flat. It might be more desirable to optimise the pulse risetime, yielding somewhat less FIR power with considerably less pump energy.

As an example we discuss a FIR-D_2O laser at 385μm. The small signal gain and saturation intensity have been measured by Drozdowicz et al.[10], for off-resonant pumping. Converted to resonant pumping the values are $g_f^{ss} = 0.36\ cm^{-1}$ and $I_{sat} = 6.7\ W/cm^2\ torr^2$. We assume availability of a pump pulse of 50 ns risetime (t_r) and adjustable power. Within the 50 ns we attempt to pump the laser to 90% of maximum FIR power. Note that according to Eq. (11) 100% of maximum power is only achieved after an infinitely long time. Hence the last term in brackets in Eq. (11) is set to 0.9 and can be written in the form $\ell = \frac{ct_r}{2} \frac{\ln R_1R_2}{\ln 0.1}$. With mirror reflectivities of R_1 = 75% and R_2 = 95% (in this value we may include other nonreflective losses) a resonator length of ℓ = 110 cm is calculated and 90% of maximum power is thus obtained after 6.8 round-trip times. From Eq. (11) the maximum extracted FIR power can now be calculated to be 17 times saturation intensity. At a pressure of 3 torr and with a beam diameter of 10 cm one obtains from the measured saturation intensity a power of 80 kW. This is valid for optimum pumping conditions. If the pump pulse is attenuated or boosted, the FIR output changes roughly according to Fig. 2.

In a similar manner to our previous examinations of a lossless resonator, the buildup of FIR radiation in an oscillator with output coupling will be studied with pump pulses in the form of linear ramps. Here we resort to numerical techniques which permits the use of the gain coefficient given by Eq. (2) instead of the approximate form of Eq. (3). FIR radiation will start to build up in a resonator provided that the small signal gain per roundtrip exceeds the total resonator losses. As both the pump and FIR intensities grow, the gain begins to saturate and eventually exactly equals the losses. The FIR power has now reached a maximum. It will decrease beyond this point even if the pump power is still growing. In Fig. 3 we display as a function of the slope of the pump pulse the following computed parameters : (a) the maximum FIR intensity, (b) the total energy conversion efficiency up to maximum FIR power, (c) the time to maximum FIR power and (d) a figure of merit defined as the ratio of maximum FIR power to pump energy consumed. If time is of no concern, it is obviously best to operate with slowly-rising pump pulses (small slope angles) to maximise both the FIR power and the energy conversion efficiency. However, less energy is consumed to achieve essentially the same FIR power in considerably shorter times by using faster rising pump pulses. Depending on the restrictions of the experimental system, optimization can be achieved with respect to certain parameters by means of Fig. 3. The fixed parameters in this figure are $R_1 = 0.8$, $R_2 = 0.95$ and hence $\alpha = -0.14$, $g_f^{ss} = 1000$. In the highly saturated regime new (primed) parameters are obtained by means of the following scaling laws :

slope	:	$\beta' = \beta \cdot \left(g_f^{ss}\right)' / g_f^{ss}$
time scale	:	$t' = t \cdot \alpha/\alpha'$
FIR power density	:	$P_f' = P_f \left(g_f^{ss}\right)' / g_f^{ss} \cdot \alpha/\alpha' \cdot r'/r$
energy conversion efficiency	:	$\varepsilon' = \varepsilon \; r'/r$
figure or merit	:	$F' = F \cdot \alpha'/\alpha \cdot r'/r$
with		$r'/r = (1-R_1')/(1-R_1) \cdot (1+R_1)/(1+R_1')$

With the help of these scaling laws and Fig. 3 it is now possible to optimise an oscillator. A linear ramp is a reasonable approximation for many types of real pump pulses. An important factor to optimise the output coupling is $(\frac{1-R_1}{1+R_1})/\alpha = 2(\frac{R_1-1}{1+R_1})/\ln(R_1R_2)$. Evenly-distributed losses in the resonator are included in α, whereas localised losses, e.g. due to windows, are included in R_2. Neglecting distributed losses, we compute the optimum reflectivity of the output coupler (assumed lossless) as function of the total localised losses $(1-R_2)$. The results are given in Table I.

5. Conclusions

We have studied the influence of pump and resonator parameters on the performance of an optically-pumped FIR laser oscillator. For resonant pumping it is found that the reflectivity of the output coupler is not a critical parameter. However, it is most important to choose an optimal pump pulse. It is especially detrimental to use pump pulses with rise times exceeding optimum. The use of too much pump power leads to spectral splitting of the gain line with corresponding decrease of the gain on line center. This is counterbalanced by power broadening at high FIR intensities. It is found that an optimum gain is obtained if pump and FIR intensities are equal multiples of their respective saturation intensities.

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TABLE I

The optimum reflectivity of a FIR oscillator output coupler as function of total localized losses (excluding output coupling). Distributed losses, such as absorption in unpumped gas, are neglected.

total localized losses	optimum reflectivity of output coupler
2%	61.1%
4	53.7
6	49.0
8	45.6
10	42.8
12	40.5
14	38.6
16	36.8
18	35.3
20	33.9

Figure Captions

Fig. 1a : The FIR power extractable per unit distance of propagation $\frac{dP_f}{dx}$ on line center for resonant pumping plotted against pump power for a variety of FIR intensities. Dimensionless units are obtained by expressing intensity in terms of saturation intensity and gain in terms of small signal gain.

Fig. 1b : The contribution of Raman and laser components to the total gain shown in Fig. 1a.

Fig. 2 : The rate of change of FIR intensity (γ) versus the rate of change of the intensity of a linear ramp type pump pulse (β) in a lossless oscillator driven into saturation.

Fig. 3 : Four optimisable parameters of a FIR pulse obtained in an oscillator pumped by a linear ramp type pump pulse. Shown as function of the slope of the pump pulse are :

- the energy conversion $\varepsilon = P_f/P_p$
- the maximum FIR intensity P_f^{max}
- the FIR pulse risetime t_r
- a figure of merit F defined in text

Fixed parameters are $R_1 = 0.8$, $R_2 = 0.95$, $g_f^{ss} = 1000$.

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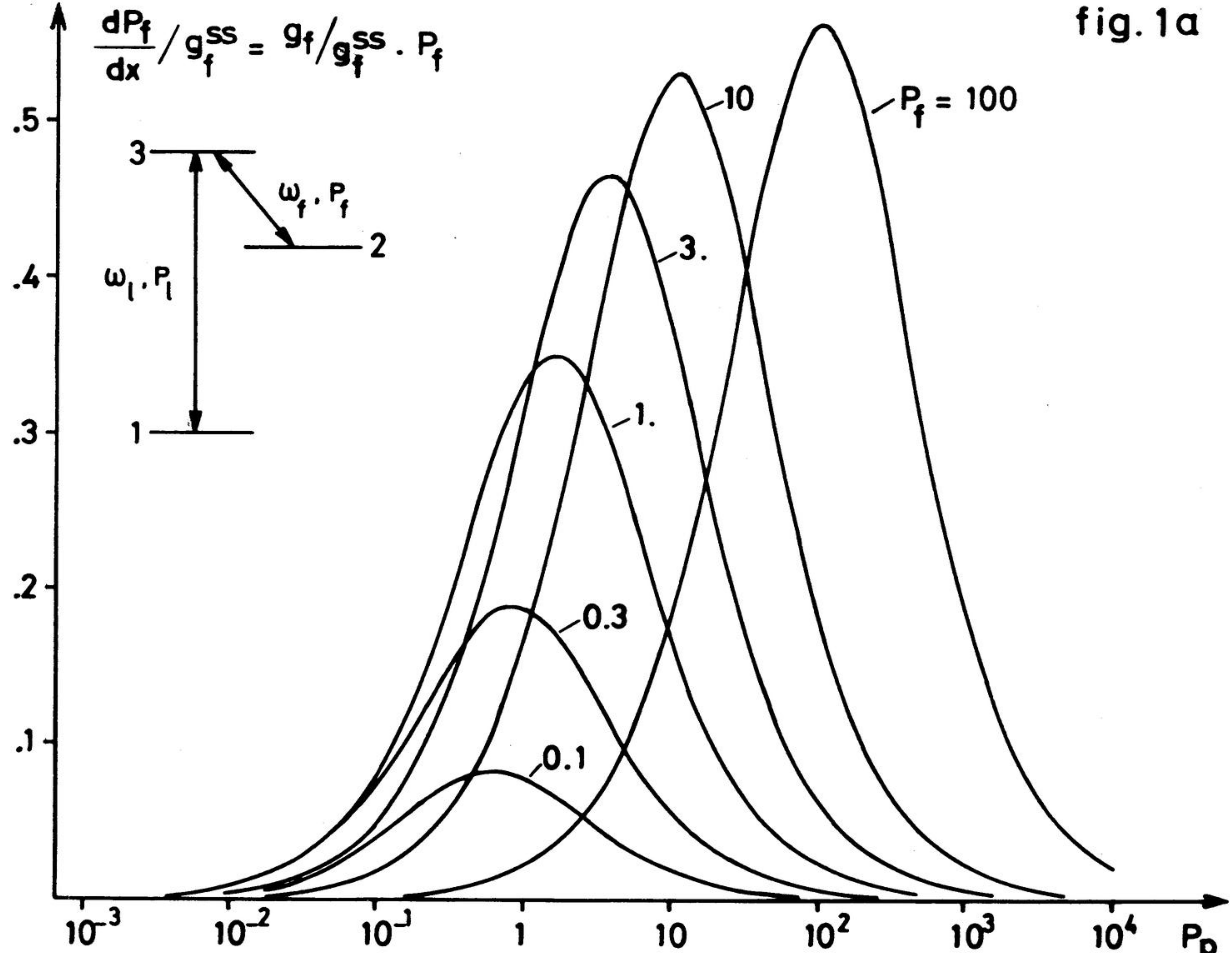
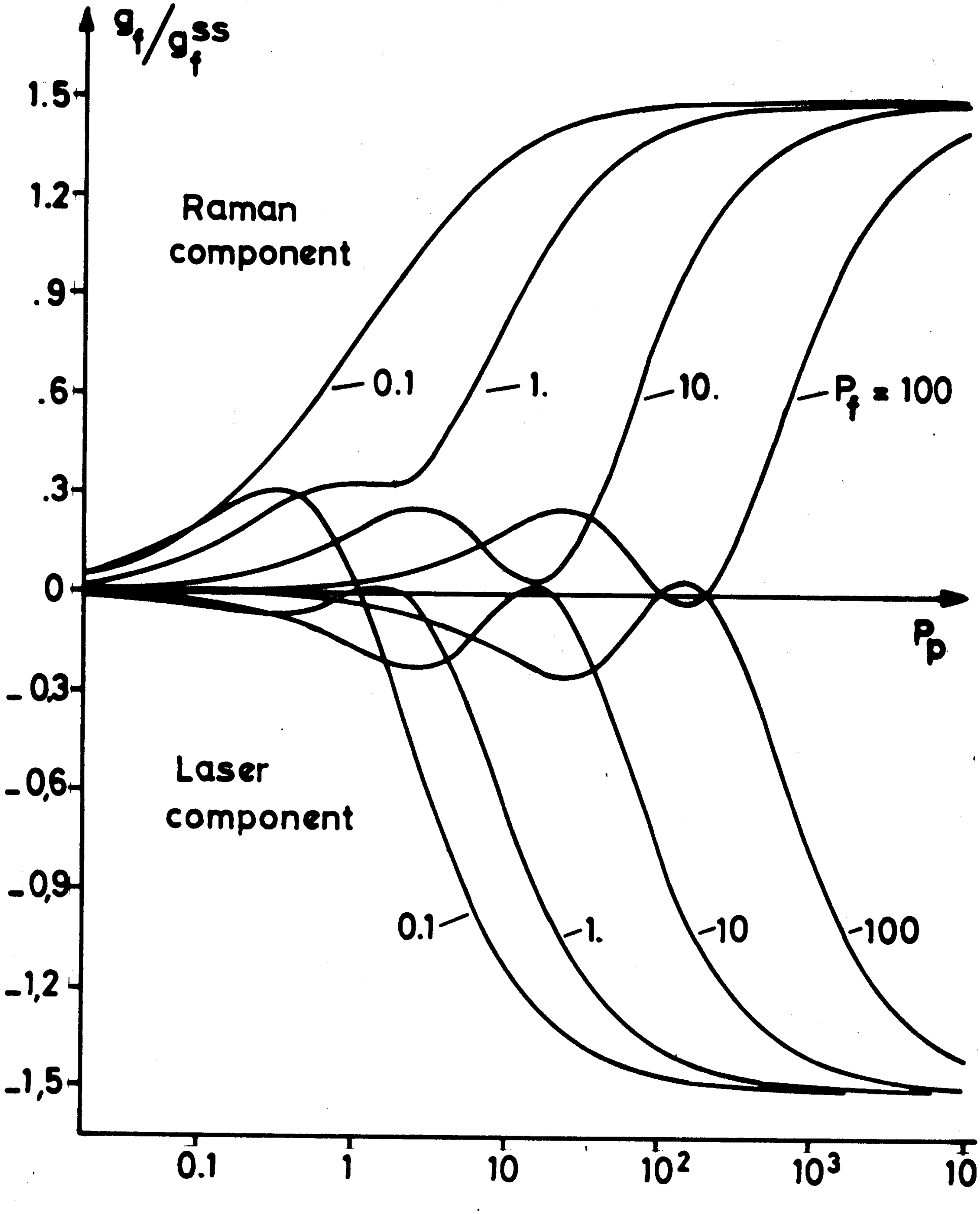


dP_f/dx / g_f^ss = g_f/g_f^ss . P_f
.5
.4
.3
.2
.1
3
2
1
ω_f , P_f
ω_l , P_l
P_f = 100
10
3.
1.
0.3
0.1
10^-3
10^-2
10^-1
1
10
10^2
10^3
10^4
P_p

fig. 1a

fig. 1b



g_f/g_f^{ss}
1.5
1.2
.9
.6
.3
0
-0,3
-0,6
-0,9
-1,2
-1,5
Raman component
Laser component
0.1
1.
10.
P_f = 100
0.1
1.
10
100
P_p
0.1
1
10
10^2
10^3
10^4

fig.2

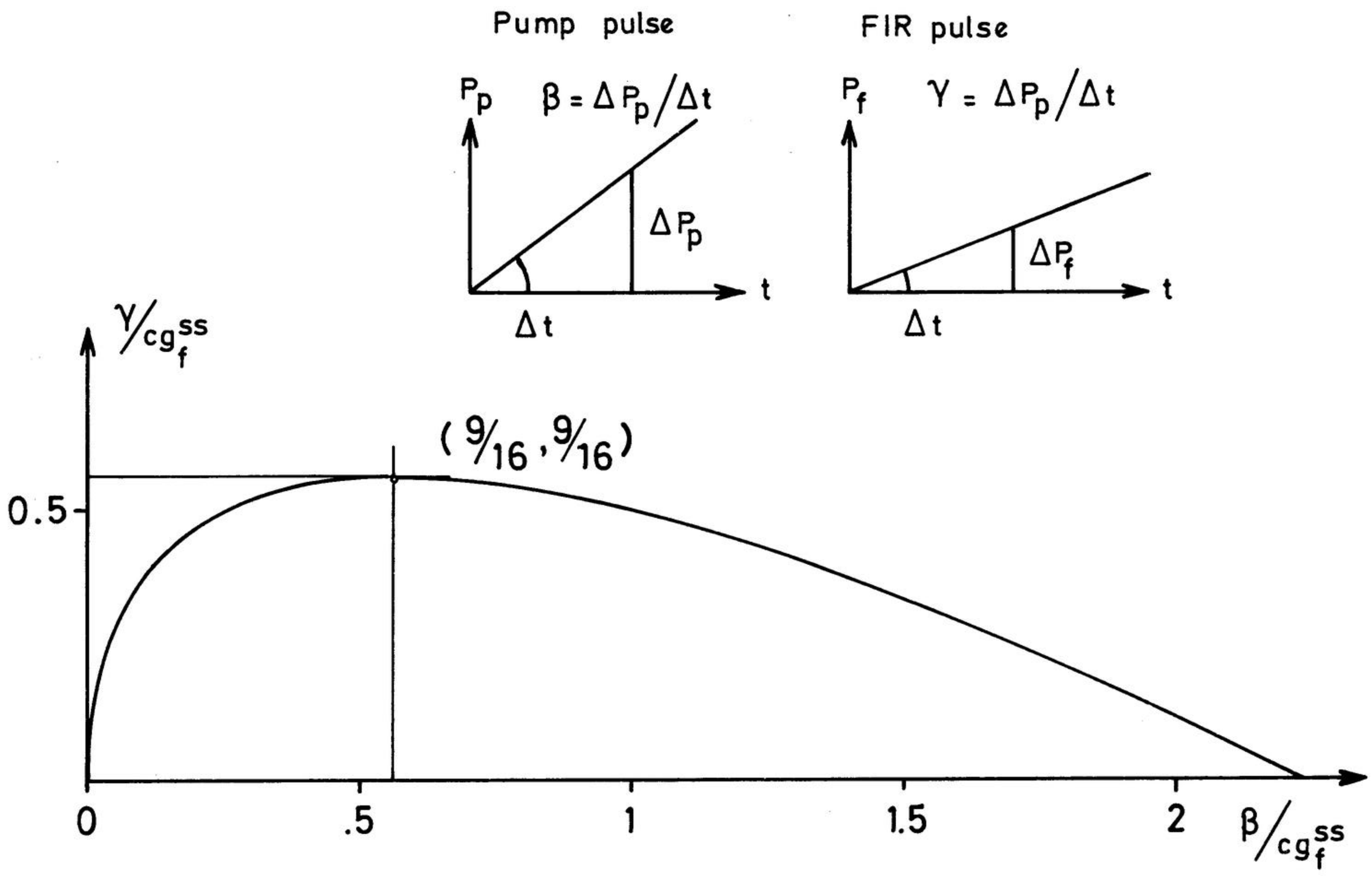
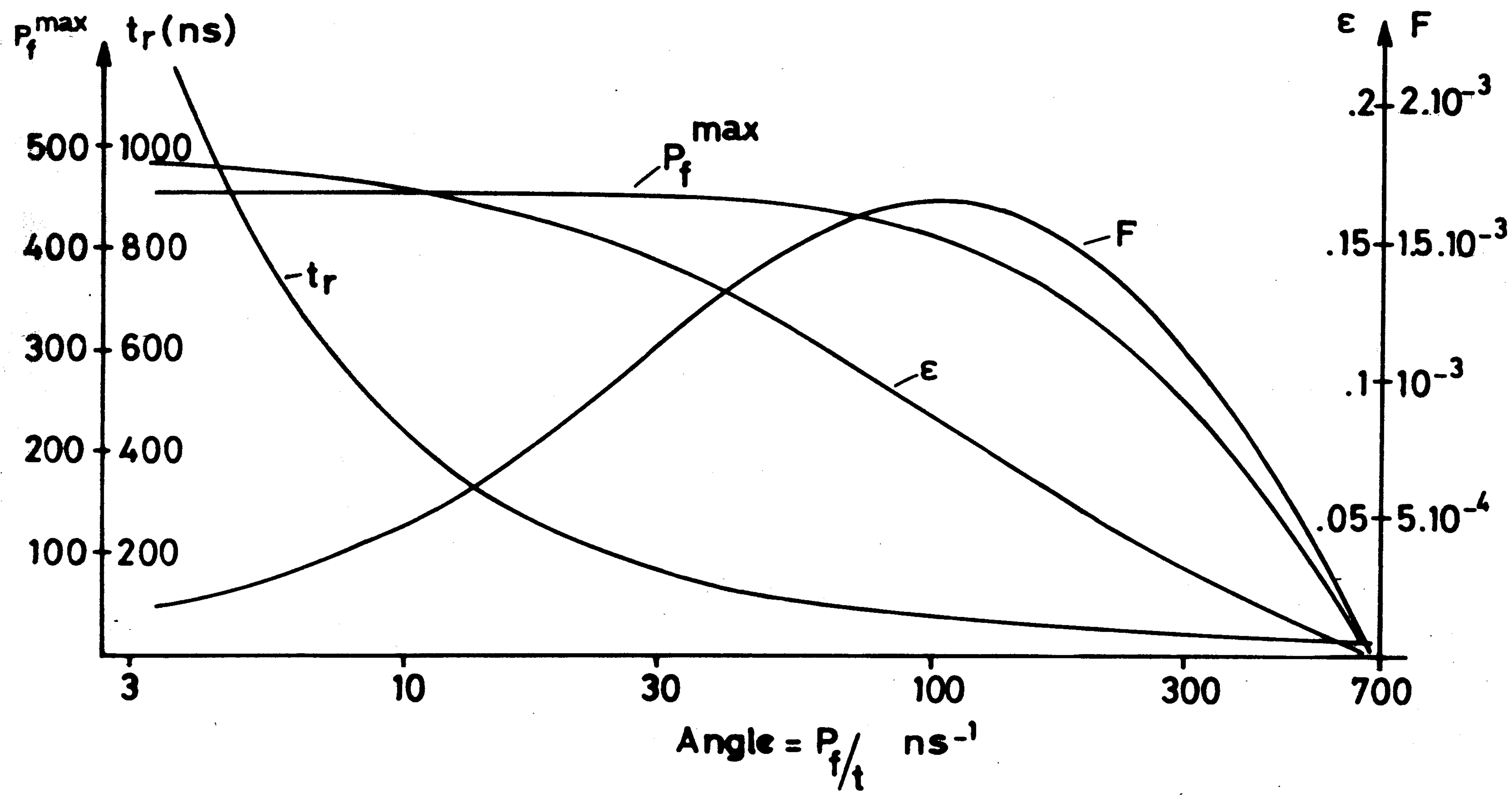

Pump pulse
FIR pulse
P_p
$\beta = \Delta P_p / \Delta t$
ΔP_p
Δt
t
P_f
$\gamma = \Delta P_p / \Delta t$
ΔP_f
Δt
t
γ / cg_f^{ss}
0.5
$(9/16, 9/16)$
0
.5
1
1.5
2
β / cg_f^{ss}

fig.3



P_f^max
t_r (ns)
500
1000
400
800
300
600
200
400
100
200
ε
F
.2
2.10^-3
.15
1.5.10^-3
.1
10^-3
.05
5.10^-4
t_r
P_f^max
ε
F
3
10
30
100
300
700
Angle = P_f/t ns^-1