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E ARMANDILLO
I J SPALDING

CULHAM LABORATORY
Abingdon Oxfordshire

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E Armandillo and I J Spalding

Culham Laboratory, Abingdon, Oxon. OX14 3DB.
(Euratom/UKAEA Fusion Association)

ABSTRACT

We have calculated the optical energy-storage and extraction capabilities of a variety of $He/N_2/CO2$ gas mixtures suitable for use in short-pulse (10^{-9}-$5x10^{-7}$s) electron-beam-preionized CO_2 laser amplifiers. For such pulse durations, which are of considerable practical interest in current laser-plasma research, a 3:1:2 gas mixture appears to be particularly useful.

The saturating, non-linear, amplifier response is shown to be well characterized by three parameters, which are related to the rotational, intra-mode and inter molecular relaxation rates. At electrical field strengths (E/N) and input energies of ~ 19 Td and ~ 200 J/litre atm. respectively, the extractable energy is typically 5-50 J/litre atm., depending upon the input pulse duration and bandwidth.

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1. INTRODUCTION

Contemporary CO_2 laser-compression (Boyer 1972) and heating (Vlases et al 1974, Donaldson et al 1975) experiments typically require pulses of order 1 kJ in energy to be delivered to a target on time scales of 1 ns - 1 μs. The shorter times are characteristic of inertially-confined solid-state target experiments, and the larger time-scales are typical of magnetically-confined plasma-heating investigations. Practical considerations (maximum available window dimensions, pre-lasing in excessively long high-gain systems, required amplifier band-width, etc.) dictate that the energy-amplifier should operate at CO_2 pressures of order 1 standard atmosphere (or more). The use of electron-beam (eb) preionization simplifies the design of such amplifiers, since the sustainer electric field can be chosen to optimize the transfer efficiency between the electrons and the $N_2(v=1)$ and CO_2 $(00v_3)$ modes, completely independently of the total sustainer (pump) current. Further optimization of the system can then be achieved by manipulating the lasing gas mixture to maximize either

(a) the over-all laser efficiency (ηe = laser power/electrical input power);

(b) the energy extractable per unit volume (ϵ_τ) by a pulse of duration τ; or

(c) the energy extractable from a given capacitor bank and discharge chamber. (Technological and cost constraints are usually sufficiently different at various laboratories that different solutions are necessary in pursuing objectives (a), (b) or (c)).

In this context several $He/N_2/CO_2$ gas mixtures have been compared computationally in the range of E/N typical of contemporary eb devices (15-25 Td). CO_2-rich gas mixtures having (volumetric) compositions of $3:\frac{1}{4}:1$, 3:1:2, and 3:2:1 have been examined in detail, and the mixture 3:1:2 at atmospheric pressure is found to combine the important advantages of high energy density storage, high optical extraction efficiency (for pulses of duration < 1 μs), and high overall electrical efficiency.

2. THE AMPLIFIER AS AN ENERGY STORAGE DEVICE

The energy exchange processes within each important molecular vibrational mode ($k_{vv} \gtrsim 10^6$ torr^{-1}sec^{-1}) are usually so fast compared to the pumping rates that a Boltzmann distribution is established and maintained throughout the pumping phase (Burak et al 1973). This fact permits one to assign vibrational temperatures T_1, T_2, T_3 to the symmetric stretch, bending and asymmetric stretch modes of the CO_2 molecule, and to the N_2 vibrational temperature. Hence by calculating the time-history of these vibrational temperatures, plus the translational gas temperature, one can calculate the level populations, the small-signal gain (α_o) and a characteristic energy:†

$$\epsilon_s = \hbar\omega\,(N_{001} - N_{100})/2 \quad . \qquad \ldots(1)$$

This so-called four-temperature-kinetic model requires an accurate solution of the Boltzmann equation for the electrons in the electrical discharge, in order to provide the electron-molecular excitation rates and drift velocities which are an essential input to the model (Lowke et al 1973, Lakshminarasimha et al 1976). It has been fully discussed by Manes and Seguin (1972), Harrach and Einwohner (1973), among others, and requires no elaboration here.

Once the level populations have been calculated in this way, the value of α_o (at the centre of the lasing line) follows from the relation

$$\alpha_o = (\lambda_j^2/4\pi^2)(n_2 - g_2 n_1/g_1)(A_{21}/B_w) \qquad \ldots(2)$$

where n_1, n_2 (and N_1, N_2) represent the rotational (and vibrational) population densities of the upper and lower levels respectively. The spontaneous emission rate for the rotation/vibration transition $2 \rightarrow 1$ is given by *

$$A_{21} = (16\pi^3/hc\epsilon\lambda_j^3)\left|\frac{\mu_{12}}{3}\right|_v^2 \left|\frac{\mu_{12}}{g_2}\right|_{rot}^2 \qquad \ldots(3)$$

where $|\mu_{12}|_v$ and $|\mu_{12}|_{rot}$ are the vibrational and rotational components

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† ϵ_s is often called 'the stored optical energy', but we shall see later that this is a misleading definition.

* Here, ϵ is the gas permitivity.

of the dipole momentum associated with the 2 → 1 transition (Cousin et al 1969). g_1 and g_2 are the degeneracy of levels 1 and 2. The line-width B_w is given by (Abrams 1974)

$$B_w = (7.61\ P_{CO_2} + 5.58\ P_{N_2} + 4.88\ P_{He}) \times 10^6 \text{ Hz torr}^{-1} \quad . \qquad ...(4)$$

The quantities ϵ_s and α_o have been evaluated as a function of E/N and the electrical input energy into the gas for the following mixtures:- 8:1:1, 3:1:1, 1:¼:1, 3:¼:1, 3:1:2, 3:2:1. (To reduce the number of variables, a constant-current electron-beam pulse, of duration 3.2 μs was postulated for these calculations. The eb duration and intensity was thus characteristic of contemporary cold-cathode electron guns. The sustainer input energy was evaluated up to the time τ_ϵ at which ϵ_s attains its maximum value.) Of the various mixtures analyzed the volumetric ratio 3:1:2 appeared to offer the best performance, i.e. it gave the highest value of ϵ_s and α_o for a given input energy. ϵ_s is of particular relevance when assessing saturated long-pulse performance, and α_o when assessing nanosecond pulse extraction.

Table 1 summarizes some characteristics of the 3:¼:1,† 3:1:2 and 3:2:1 gas mixtures, whilst Figs.1-3 illustrate the variation of ϵ_s and α_o with input energy at E/N values of 15, 19 and 21 Townsends respectively, ($1\text{Td} = 10^{-17}\text{V cm}^{+2}$). τ_ϵ and the time at which the maximum value of α_o occurs, τ_α, are also plotted. A knowledge of τ_ϵ and τ_α is necessary, of course, in order to optimize the relative timing between a driving oscillator pulse and the amplifier pumping pulse. Figs.1-3 indicate that both α_o and ϵ_s increase rapidly with electrical discharge energy at low pumping levels (< 200J/la)*; this includes a region of high electrical efficiency. As the pumping rate is further increased, first α_o and then ϵ_s saturate; finally at pumping rates $\gtrsim$ 300J/la they both decrease monotonically, resulting in lower electrical efficiencies. This decrease occurs because of thermal population of the lower laser level and distribution of energy between several states in the higher anti-symmetric stretch mode (see, for example, Orishich and Ponomarenko 1975).

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† Leland and Kircher (1975) analysed the performance of a 3:¼:1 gas mixture, but assumed a fixed gas temperature during the discharge.

* 1 la ≡ 1 litre.atmosphere.

Because of the severe increase in T_1 and T_2 with input energy, τ_ϵ and τ_α monotonically decrease with input energy. Indeed, for the conditions illustrated in Figs.1-3 the maximum values of α_o and ϵ_s both occur before the end of the eb current pulse (3.2 μs) when the pumping exceeds ~ 200J/la. (Note that $\tau_\epsilon > \tau_\alpha$ for any input energy, and for any of the gas mixtures discussed, for the same reason that α_o saturates with energy input before ϵ_s.)

3. THE AMPLIFIER AS AN ENERGY DELIVERING DEVICE

The energy density (J/la) which can actually be extracted from an amplifier depends not only on the input pulse intensity, but also on its pulse duration, so that ϵ_s does not always characterize the amplifier performance in a meaningful way. The extractable energy can be greater or less than ϵ_s, depending upon the relative rate of variation of the laser intensity and the various energy-exchange rates within the amplifying medium.

To illustrate this in a quantitative fashion we have calculated the maximum energy which can be extracted from a 3:1:2 gas mixture, for a wide range of laser pulse-durations. The electrical energy input up to time τ_α is chosen to be 197 J/la, at a mean sustainer field E/N of 19 Townsends; as we saw in section 2, these conditions represent a good compromise between the practical requirements of high-density storage of optical energy, and high over-all electrical efficiency. With initial conditions $T_1 = T_2 = T_3 = T_{N_2} = T_{gas}$ (translational) = 293°K, an eb pulse of duration 3.2 μs raises the modal temperatures to the following values at the time τ_α: $T_1 \sim T_2 = 451$°K, $T_3 = 1761$°K, $T_{N_2} = 1808$°K, with $T_{gas} = 378$°K. It follows from equations 2 and 1 that α_o(P(20),10.4 μm band) = 5% cm^{-1}, and ϵ_s = 6.8 J/la.

The total energy stored in the multiplicity of levels within each of the vibrational modes can also be calculated as follows, in the harmonic oscillator approximation:

$$E_{\nu_i} = h\nu_i N_{CO_2} \left[\frac{\exp(-h\nu_i/kT_i)}{1 - \exp(-h\nu_i/kT_i)} \right] \qquad \ldots(5)$$

The (total) energy stored in each of the symmetric stretch (i=1), bending (i=2) and asymmetric stretch (i=3) modes is thus

$$\left.\begin{aligned} E_{\nu_1} &= 3.3 \text{ J/la} \\ E_{\nu_2} &= 29.9 \text{ J/la} \\ E_{\nu_3} &= 66.9 \text{ J/la} \end{aligned}\right\} . \qquad \ldots(6)$$

An identical equation to (5) can be written for the sum of energies stored in the various levels of the nitrogen vibrational mode, giving

$$E_{N_2} = 35.8 \text{ J/la} . \qquad \ldots(7)$$

Thus the total electrical energy converted by electron collisions into molecular vibrational excitation is $E_{\nu_1} + E_{\nu_2} + E_{\nu_3} + E_{N_2} = 136$ J/la.

During the pumping process the gas temperature rises from 293°K to 378°K, due to non-resonant processes and molecular relaxations; the energy loss corresponding to this temperature rise is 58.7 J/la, giving a total conversion of electrical into vibrational energy of ~ 99%. The remaining 1% of the electrical pumping energy is lost by low energy electrons making elastic collisions, and in electronic excitation and ionization.

Taking into account the quantum efficiency ($\eta_q = 0.41$) the ($\lambda = 10.6$ μm) photon energy potentially available from the CO_2 ν_3 mode becomes $0.41\ E_{\nu_3} = 27$ J/la. If one adds the energy available from the nitrogen vibrational levels, the maximum extractable $\lambda = 10.6$ μm photon energy becomes $0.41(E_{\nu_3} + E_{N_2}) = 42$ J/la.

After the optimization of the electrical excitation efficiency, it is interesting to examine the optical extraction efficiency, neglecting any geometrical losses, etc. A non-linear theory of the amplifier/light pulse interaction is necessary for this calculation, and a suitable model has therefore been developed which includes all processes of interest on time-

scales in the sub-nanosecond to μs region, (including coherent effects). The physical justification for the model has been discussed previously (Armandillo and Spalding, 1975); here we are interested only in quantitative conclusions appropriate to the gain medium discussed earlier. To be specific, let us assume an optical input intensity having a time variation of the form

$$f(x) = x^2 \exp(-x^4/2) \qquad \ldots(7)$$

where $x = (\tau/\tau_o)$, τ is reduced time $(t - z/c)$, τ_o is the time at which peak intensity occurs, and $f(0) = f'(0) = 0$. Fig.4 illustrates the shape of this input pulse, whilst Fig.5 shows the (saturated) amplifier response as a function of the input pulse duration $\tau_{\frac{1}{2}}$(FWHM). The semi-logarithmic plot in Fig.5 shows quite clearly three points of inflexion, which in the language of system theory (see for example Kuo 1962) indicate 3 positive (saturable) poles in the corresponding frequency response. These poles occur approximately at $1/\tau_R$, $1/\tau_{vv}$ and $1/\tau_{CO_2\text{-}N_2}$, i.e. at the inverse of the rotational equilibration time ($\lesssim 2 \times 10^{-10}$s atmospheres), the intramode equilibration time ($\lesssim 4 \times 10^{-9}$s atmospheres) and the CO_2- N_2 vibrational exchange time ($\lesssim 10^{-7}$s atmospheres).

For pulse durations $\lesssim$ 1ns, the optical extraction efficiency is sensitive to the number of rotational lines included within the input pulse; as indicated in Fig.5 single-line extraction (represented by the continuous trace) is relatively inefficient, whilst multi-line operation* (e.g. 3 lines of equal intensity at P_{20}, P_{22}, P_{24}) removes practically all the energy available from the 001-100 band. At slightly longer times, $\gtrsim$ 10 ns, the response flattens to ~ 27 J/la (i.e. 0.41 E_{ν_3}) because the intramode reservoir saturates and the relatively slow CO_2-N_2 exchange and pumping collisions are not yet operative. For pulse durations of ~ 100 ns all of the energy stored in the ν_3 mode is extractable, but the effect of pumping is clearly noticed for pulses longer than ~ 100 ns. Under these conditions the depleted 001 vibrational level can be replenished directly through low-energy electron collisions, and through the N_2-CO_2 channel.

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* Indicated by an asterisk in Fig.5

4. CONCLUSIONS

For many short-pulse eb preionized CO_2 laser applications a $He:N_2:CO_2$ gas mixture in the proportions 3:1:2 would appear to exhibit several advantages. Calculations indicate that these include high energy density storage, optical extraction efficiencies, and over-all laser efficiency. (A purely technological advantage which has not been discussed, but which it is convenient to note here, is the higher DC break-down strength of helium-lean gas mixtures.)

Use of a non-linear pulse-propagation code, coupled with a kinetic model for the amplifier, indicates that nano-second 10.6 μm laser pulses might be capable of extracting energy densities of up to 8 J/la, multi-line. (This energy density requires pumping energies and E/N marginally greater than those tabulated in para.3, and corresponds to an over-all efficiency of 4% at 1 standard atmosphere). At longer pulse durations which may become attractive for plasma-heating experiments, the corresponding numbers are ~ 30 J/la and 16% for 100 ns pulses, and ~ 50 J/la and 25% for 500 ns pulses. These idealized calculations may be modified, of course, by variations to the input conditions to the computer codes (e.g. inclusion of effects due to gas impurities and break-down products) and by practical complications, such as losses in optical components.

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TABLE 1 VARIATION OF α_o AND ϵ_s WITH GAS MIXTURE, FOR VARIOUS E/N AND PUMPING ENERGIES (W_{in})

	E/N = 15 Td		E/N = 19 Td		E/N = 21 Td	
W_{in} (J/la)	α_o (% cm^{-1})	ϵ_s (J/la)	α_o (% cm^{-1})	ϵ_s (J/la)	α_o (% cm^{-1})	ϵ_s (J/la)
80	(a) 2.8	3.4	3.2	3.5	3.3	4.4
	(b) 3.1	3.5	3.4	3.9	3.5	4.0
	(c) 2.1	2.3	2.3	2.5	2.6	3.1
150	(a) 3.9	5.4	4.5	5.9	4.8	6.1
	(b) 4.2	5.2	4.4	5.8	4.7	5.8
	(c) 2.5	2.9	3.3	3.8	3.5	4.0
200	(a) 4.2	6.0	5.1	6.6	5.3	6.8
	(b) 4.6	6.2	5.0	6.0	5.2	6.6
	(c) 2.8	3.4	3.7	4.6	3.9	4.3
300	(a) 4.3	6.4	5.2	7.4	5.5	7.8
	(b) 4.4	6.4	4.9	6.6	5.1	6.7
	(c) 3.3	4.5	3.9	5.2	4.1	5.5

NB Gas mixture (a) 3:1:2
(b) $3:\frac{1}{4}:1$
(c) 3:2:1

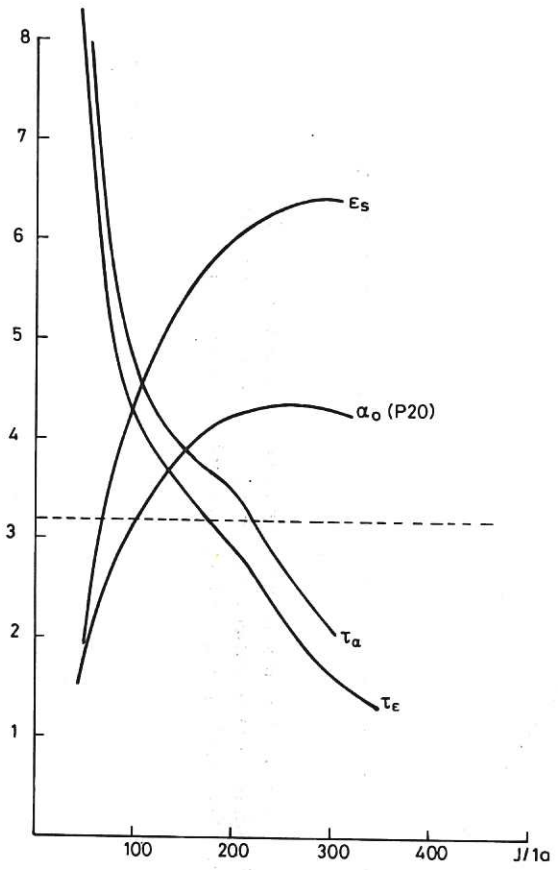

Fig.1 Variation of α_o (%cm^{-1}), ϵ_s (J/la), τ_ϵ (μs) and τ_α (μs) as a function of electrical input into the 3:1:2 gas mixture. (The dashed line indicates the 3.2 μs eb current pulse-duration, for comparison with τ_ϵ and τ_α) E/N = 15 Td.

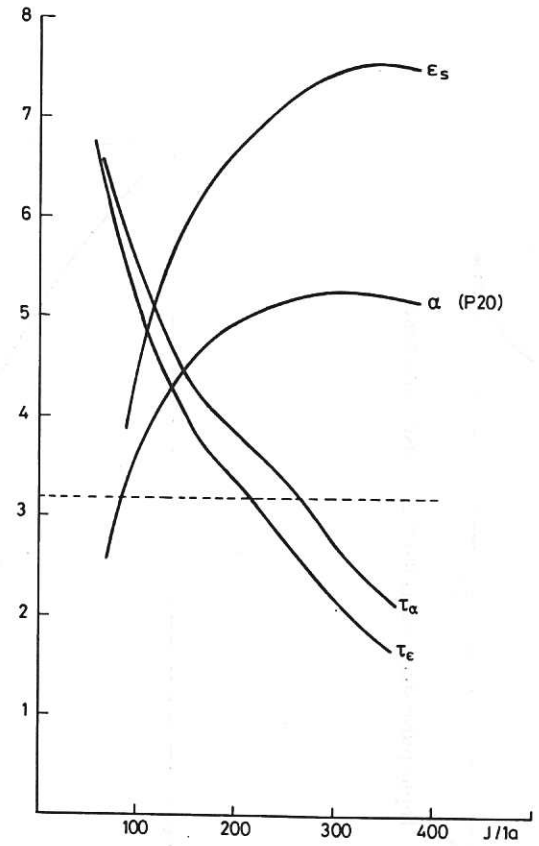

Fig.2 As Fig.1; E/N = 19 Td.

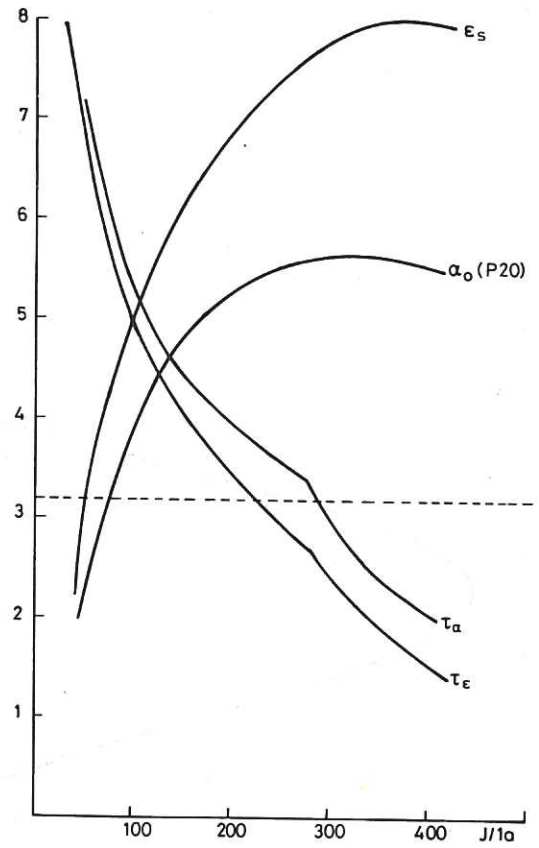

Fig.3 As Fig.1; E/N = 21 Td.

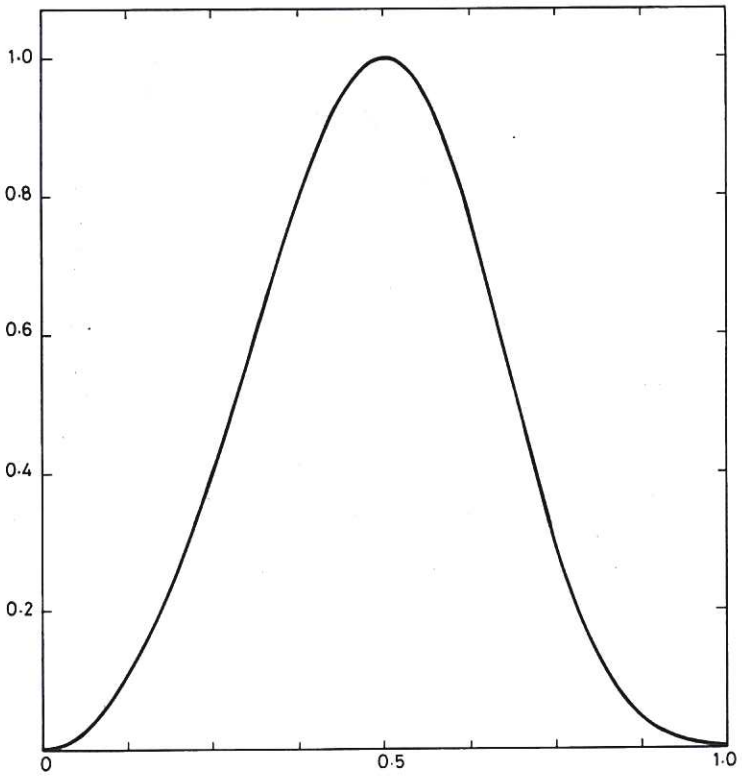

Fig.4 Input pulse shape (normalized intensity and time).

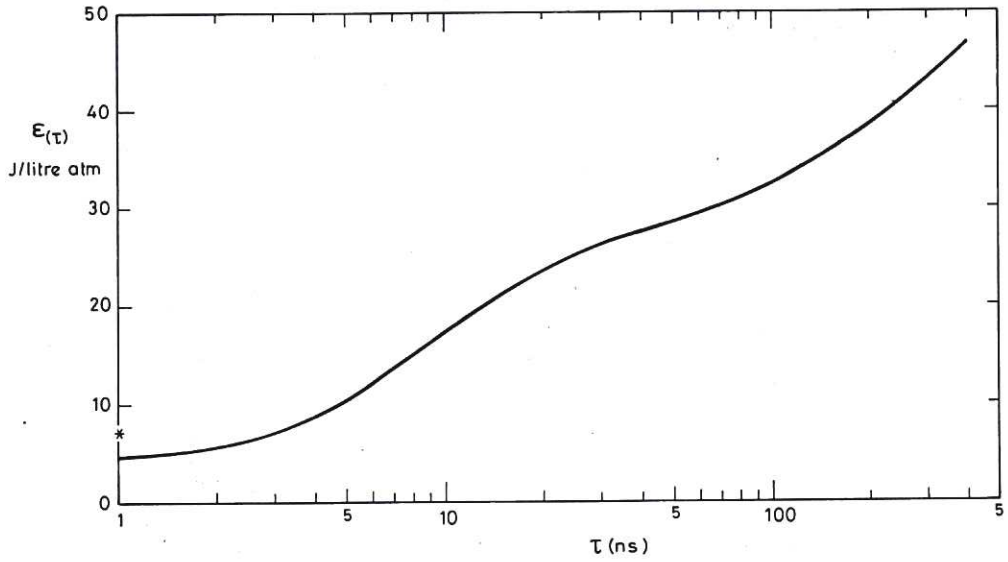

Fig.5 Energy extractable, in joules per litre atmosphere, as a function of the FWHM pulse duration. (The amplifier is that illustrated in Fig.2, operated at 1 standard atmosphere, with an electrical input of 197 J/la at time τ_a).