IX. ELECTRODYNAMICS OF MEDIA^{*}

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A. GENERATION AND AMPLIFICATION OF VERY SHORT CO₂ LASER PULSES

Since its discovery five years ago, the CO_2 laser has generated a great deal of interest because of its high efficiency (~17%), high power, and ease of construction. Moreover, the CO_2 laser may be Q-switched because of its high gain and the long lifetime of the upper laser level. These intense pulses are of interest because an atmospheric window exists at the CO_2 10.6- μ wavelength, which makes a laser radar an attractive possibility. Furthermore, short pulses with high peak powers make second-harmonic generation to the near infrared attractive.

We report here on a novel scheme for generating short CO_2 laser pulses and some studies on the amplification of these pulses. This method is unique because it permits generation of pulses with rise times not limited by the inverse bandwidth of the CO_2 laser medium (~100 MHz). The amplifier studies indicate that we can obtain undistorted amplification at frequencies greater than the inverse bandwidth.

Conventional means of Q-switching involve use of a rotating mirror and the introduction of SF_6 , a saturable absorber, into the cavity. Neither of these methods is satisfactory for generating single pulses with rise times shorter than the inverse bandwidth.

The method that we use is cavity dumping with a Ga As electro-optic modulator. Figure IX-1 is a schematic diagram of the equipment which includes the amplifier. The Q-switched oscillator is comprised of a curved mirror, 3.5 m of gain tubes, a Ge polarizer, the Ga As electro-optic modulator, and a partially transmitting Irtran IV flat that completes the cavity. When no voltage is applied to the Ga As modulator, the laser will oscillate cw with polarization in the plane of the paper. When a voltage is applied to the Ga As modulator to make it into a quarter-wave plate, the cw oscillation stops because of the loss introduced by the Ge plate. Now if the quarter-wave voltage is dropped to zero, the oscillation in the cavity, monitored by detector A, builds up from

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Fig. IX-1. Experimental arrangement.

noise. Approximately 300 nsec later, when the Q-switch power is at its peak, the quarter-wave voltage is reapplied to the Ga As modulator. This causes the polarization of the field inside the cavity to flip 90° and be dumped off the Ge plate (Ge has an index of refraction of 4, thereby causing an 80% reflection per face of radiation polarized perpendicular to the Brewster angle). The output pulse is as long as the round-trip transit time of the cavity (in this case ~48 nsec) and has a rise time limited only by the electronic switching time of Ga As, which in this case is 5 nsec. This pulse is monitored by detector B; all detectors used in this experiment are Ge:Cu photo-conductive detectors with a measured response time of less than 1 nsec. The pulse is then amplified in a multipass amplifier which gives a total of 5 m of amplification.¹ In the lower part of Fig. IX-1 there are shown typical oscilloscope traces for the quarter-wave voltage and detector responses. Note that the intensity inside the cavity (det. A) drops to zero when the pulse is dumped.

Figure IX-2 is an oscilloscope photograph of the intensity inside the cavity (upper trace) and the voltage on the Ga As crystal (lower trace). Figure IX-3 shows the dumped pulse (upper trace) and intensity inside the cavity (lower trace). The rise time of the pulse is approximately 5 nsec, and its peak power is approximately 5 kW. Figure IX-4 is a multiple exposure of Fig. IX-2, but during one Q-switching sequence the voltage was not reapplied to the Ga As. On that one occasion, the intensity in the cavity leaked out through the IR IV mirror; this is evident in the upper trace. Figure IX-5 is like Fig. IX-2, except that proper axial tuning of the cavity has led to spontaneous mode-locking behavior. The ringing of the upper trace is $2\ell/c$, and the



Fig. IX-2.

Upper trace: intensity inside the cavity. Lower trace: GaAs switching voltage, 2 kV/major division. 100 nsec/major division.



Fig. IX-3.

Upper trace: dumped pulse, 5 nsec rise time, 5 kW peak power. Lower trace: Intensity inside the cavity. 100 nsec/major division.



Fig. IX-4.

Multiple exposure of Q-switching action. 100 nsec/major division.



Fig. IX-5. Spontaneous mode locking.

100 nsec/major division.



Fig. IX-6. Amplification of short, intense pulse. Upper trace: amplified pulse. 10 nsec/major division. Lower trace: input pulse.



Fig. IX-7. Amplification of short, nonsaturating pulse. Upper trace: input pulse. Lower trace: amplified pulse. 10 nsec/major division.



Fig. IX-8. Pulse sharpening and observation of nutation effect. Upper trace: Amplified pulse. Lower trace: Input pulse. 10 nsec/major division.

width of the pulses indicates that only 3 modes are being locked. Further investigations are under way to induce mode locking.

1. Amplification of Short Pulses

Given the fact that we have generated pulses that are not bandwidth-limited, how do we now set about amplifying them so that they will not be bandwidth-limited? This can be done if the pulse input rises quickly enough and is intense enough that the rate of induced transitions exceeds the inverse bandwidth, that is, $\mu E/\hbar > 1/\Delta \nu$. Another way of phrasing it is that we must get sufficient energy into the amplifying medium in a time comparable to or shorter than the inverse bandwidth. Note that in this regime, the amplification process cannot be described solely by rate equations, but rather density matrix equations are necessary. A full study of this amplification process shows that if the input pulse is strong enough, it can turn an initially amplifying medium into an absorber that causes pulse sharpening.², ³

Some preliminary experimental results are shown here. Figure IX-6 shows a pulse before and after amplification. The lower trace is det. B and the upper trace, the amplified pulse, is det. C. Note that the rise time of the amplified pulse is ~15 nsec. Similar data have been taken at the same pressures (~11 Torr) for an input pulse of extremely low intensity (milliwatts). This is shown in Fig. IX-7. The rise time of the input pulse (upper trace) is less than 1 nsec, while the amplified pulse has a rise time of 25 nsec. So the intense pulse can be amplified with a rise time faster than the inverse bandwidth. Figure IX-8 shows an example of pulse sharpening at pressures less than 1 Torr. The lower trace is the input pulse, and the upper trace is the amplified pulse. Since the peak amplification is only 10%, the pulse is definitely absorbed on the tail end. This is the first direct observation of a "nutation" effect in a CO₂ laser. Figure IX-9 shows a computer calculation of such an effect. It is surprising that we were able to see such an effect at all



Fig. IX-9. Computer calculation.

79

(IX. ELECTRODYNAMICS OF MEDIA)

because (i) in the computer calculation a two-level system was assumed, and (ii) a planewave interaction with the medium was assumed. The first point is not valid, since if our intense interaction begins in a time short compared with an inverse bandwidth, that is, before one collision time, the medium is degenerate. There exist different dipole moments

$$\mu_{j} = \mu \sqrt{(J+1)^{2} - M^{2}}$$

that interact with the electromagnetic field. If the CO_2 laser is operating in the P(20) transition, 21 different dipole moments exist that "ring" at slightly different rates; our detector averages them all and tends to smear out any pulse sharpening.

The second point is quite similar. If the pulse has a Gaussian cross section, the intense center of the beam interacts strongly, thereby causing a deep nutation, whereas the edges may not ring at all. Again our detector averages all of this, thereby making it difficult to see any pulse sharpening.

P. W. Hoff, H. A. Haus

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B. CROSS RELAXATION IN A MULTILEVEL SYSTEM

In a previous report,¹ we presented a theory of the effect of cross relaxation on the Lamb dip in a two-level system. Our main intention in developing such a theory was to cover the case of the CO_2 system operating at 10.6 μ , which cannot be characterized by a simple two-level system. In the present report, we shall present a sequel to this investigation. We shall give an analysis of a multilevel system and then show how the results presented here lead to a description of the Lamb dip observed in a multilevel system in terms of an equivalent two-level system. In fact, the rate equations in the steady state of a multilevel system, only a pair of levels of which interacts with a laser field, can always be reduced formally to those of a two-level system by simply eliminating the population densities of the nonlasing levels from the expression for the population densities of the lasing levels. The resulting parameters are then expressible in

terms of the determinants of the coefficients of the complete rate equations. We shall simplify the multilevel system equations sufficiently that a closed-form solution can be obtained. Since in the CO_2 laser systems the relaxation times of the lower rotational vibrational levels are faster than those of the upper levels, a simplifying approximation will be made, in which the population in the lower levels is disregarded entirely. Actually, the methods developed here are applicable to the more realistic system of upper and lower levels, with the result that a fourth-order determinant has to be solved instead of a second-order one.

We shall assume that three relaxation processes take place in the system of the upper levels: (a) fast rotational level relaxation; (b) slower cross relaxation through the Doppler profile of each level; and (c) relaxation out of the levels under consideration into all of the other levels.

The rate equation for the $j^{\mbox{th}}$ level with its velocity-dependent population density $n_j(v)$ is

$$\dot{n}_{j}(v) = -\gamma_{j}(v) n_{j}(v) - \int \Gamma_{j}(v', v) n_{j}(v) dv' + \int \Gamma_{j}(v, v') n_{j}(v') dv' - \sum_{k \neq j} \Gamma_{kj}(v) n_{j}(v) + \sum_{k \neq j} \Gamma_{jk}(v) n_{k}(v) + R_{j}(v) - W(v) n_{j}(v) \delta_{j\ell}.$$
(1)

The ℓ^{th} level is assumed to be the lasing level. Note that we assume that the relaxation from the jth level to the kth level occurs only within a particular velocity group v, v+dv. The cross relaxation through the velocity profile is described by the terms $\int \Gamma_j(v',v) n_j(v) dv'$ and $\int \Gamma_j(v,v') n_j(v') dv'$. Only the ℓ^{th} level couples to the radiation field. The function W(v) is the rate of induced emission² (cf. the previous report¹):

$$W(v) = \sum_{k,\omega} \frac{c}{h\omega} \sigma(v,k,\omega) E^{2}(k,\omega).$$
(2)

The term $R_j(v)$ represents the pump into the jth level. The population in the lower lasing level is disregarded for simplicity. A generalization to include lower level population densities is not difficult, but lengthy.

In order to obtain equations leading to closed-form solutions, we make certain reasonable assumptions. For the relaxation rate $\gamma_j(v)$ into all levels of the system not explicitly included in the rate equation we assume that it is velocity and j-independent

$$\gamma_{j}(\mathbf{v}) = \gamma. \tag{3}$$

For the cross relaxation through the Doppler profile we assume

$$\Gamma_{j}(\mathbf{v}',\mathbf{v}) \ n_{j}^{(e)}(\mathbf{v}) = \Gamma_{j}(\mathbf{v},\mathbf{v}') \ n_{j}^{(e)}(\mathbf{v}') = \frac{1}{\tau} \frac{n_{j}^{(e)}(\mathbf{v}) \ n_{k}^{(e)}(\mathbf{v})}{N_{j}^{(e)}},$$
(4)

where $n_j^{(e)}(v)$ is the equilibrium (in absence of laser field) density distribution of the j^{th} level. Here

$$N_j^{(e)} = \int dv n_j^{(e)}(v).$$

The first equality represents no more than the principle of detailed balance. The second equality is analogous to the one used previously in the treatment of the two-level system. It is reasonable and necessary to obtain a closed-form solution. The relaxation rates into all other upper levels are assumed to satisfy the relations

$$\Gamma_{kj}(v) n_{j}^{(e)}(v) = \Gamma_{jk}(v) n_{k}^{(e)}(v) = \Lambda \frac{n_{j}^{(e)}(v) n_{k}^{(e)}(v)}{\sum_{m} N_{m}^{(e)}}.$$
(5)

Note that the first equality is again the principle of detailed balance, and the second equality implies that a particle is randomized after a single collision.³ Furthermore, it implies that all levels are in a sense equivalent because only one constant, Λ , is used to describe their relaxation rate. This is not quite true to the conditions in CO₂ in which the rotational relaxation within a vibrational level proceeds at a rate faster than the relaxation among the vibrational states. To do justice to this system one would need to separate the upper levels into systems of sublevels describing the relaxation among levels of a vibrational state separately from the relaxation among levels of different vibrational states. We shall see, however, that when the relaxation rate Λ is large (our meaning of "large" will be defined more precisely) it does not enter into the final answer. In the same way, a finer subdivision than the one outlined above leads to the same conclusion if the relaxation rates are all large (the coupling among the levels is tight).

Next we consider the steady-state format of Eq. 1. We make the van der Pol assumption, in which the population density multiplying W(v) is replaced by the population density $n_{l}^{(e)}(v)$ of the lasing level in the absence of a laser field. Introducing (2) through (5), we obtain for the change in population density $n_{j}(v) - n_{j}^{(e)}(v) = \Delta n_{j}(v)$

(IX. ELECTRODYNAMICS OF MEDIA)

$$0 = -\gamma \Delta n_{j}(v) - \frac{1}{\tau} \Delta n_{j}(v)$$

$$+ \frac{1}{\tau} \frac{n_{j}^{(e)}(v)}{N_{j}^{(e)}} \int \Delta n_{j}(v') dv' - \Lambda \sum_{k \neq j} \left(\frac{N_{k}^{(e)}}{\sum m N_{m}^{(e)}}\right) \Delta n_{j}(v)$$

$$+ \Lambda \sum_{k \neq j} \frac{N_{j}^{(e)}}{\sum m N_{m}^{(e)}} \Delta n_{k}(v) - W(v) n_{j}^{(e)}(v) \delta_{j\ell}.$$
(6)

We introduce the symbol Z to stand for the equivalent number of levels

$$Z = \frac{\sum_{m}^{\sum} N_{m}^{(e)}}{N_{\ell}^{(e)}}.$$

Furthermore, we introduce the symbol f(v) for the normalized Doppler profile, which must be equal for all levels in an equilibrium situation,

$$f(v) = \frac{n_{j}^{(e)}(v)}{\int n_{j}^{(e)}(v) dv} = \frac{n_{j}^{(e)}(v)}{N_{j}^{(e)}}.$$
(7)

We assume the closed-form solutions, for the lasing level

$$\Delta n_{\ell}(v) = [A+BW(v)]N_{\ell}^{(e)} f(v), \qquad (8)$$

and for the remaining levels

$$\Delta n_{j}(v) = [C+DW(v)]N_{j}^{(e)} f(v) \qquad j \neq \ell,$$
(9)

where A, B, C, D are constants independent of the velocity, and C and D, which pertain to the nonlasing levels, are independent of the index j. When these assumed solutions are introduced into (6) for j = l, we find

$$0 = -\left[\gamma + \frac{1}{\tau} + \Lambda \left(1 - \frac{1}{Z}\right)\right] \left[A + BW(v)\right]$$
$$+ \frac{1}{\tau} \int \left[A + BW(v')\right] f(v') dv'$$
$$+ \Lambda \left(1 - \frac{1}{Z}\right) \left[C + DW(v)\right] - W(v), \qquad (10)$$

and for $j \neq l$

$$0 = -\left[\gamma + \frac{1}{\tau} + \frac{\Lambda}{Z}\right] \left[C + DW(v)\right]$$
$$+ \frac{1}{\tau} \int \left[C + DW(v')\right] f(v') dv'$$
$$+ \frac{\Lambda}{Z} \left[A + BW(v)\right].$$
(11)

To be consistent with the original assumption that the coefficients A through D are independent of v, the multipliers of W(v) in Eqs. 10 and 11 have to cancel, yielding two equations for B and D, which give:

$$B = -\frac{\left(\gamma + \frac{1}{\tau} + \frac{\Lambda}{Z}\right)}{\left(\gamma + \frac{1}{\tau}\right)\left(\gamma + \frac{1}{\tau} + \Lambda\right)}$$

$$D = \frac{\frac{\Lambda}{Z}}{\left(\gamma + \frac{1}{\tau} + \frac{\Lambda}{Z}\right)} B.$$
(12)
(12)

When (12) and (13) are used in (11) and (12) we finally obtain for the population inversion of the lasing level

$$\Delta n_{\underline{\ell}}(v) = -n_{\underline{\ell}}^{(e)}(v) \frac{\frac{\gamma + \frac{1}{\tau} + \frac{\Lambda}{Z}}{(\gamma + \frac{1}{\tau})(\gamma + \frac{1}{\tau} + \Lambda)}}{\left(\gamma + \frac{1}{\tau}\right)(\gamma + \frac{1}{\tau} + \frac{\Lambda}{Z}) + \frac{\Lambda^2}{Z}}{\frac{1}{\tau}\int W(v') f(v') d\omega'} \right].$$
(14)

Equation 14 gives the change in the inversion of the lasing level. This expression is identical in form with expression obtained for the two-level system, 4 except for a reinterpretation of the parameters in this expression. If we set

$$\gamma \tau' = \frac{\gamma(\gamma + \Lambda) \left(\gamma + \frac{1}{\tau} + \frac{\Lambda}{Z}\right) \tau}{\gamma^2 + 2\gamma \frac{\Lambda}{Z} + \frac{1}{\tau} \left(\gamma + \frac{\Lambda}{Z}\right) + \frac{\Lambda^2}{Z}}$$

we obtain for the Lamb dip of a multilevel system

$$P = K \frac{G_0 - L \exp \frac{(\omega - \omega_0)^2}{2\Delta\omega^2}}{1 + \frac{1}{1 + (\omega - \omega_0)^2 T_2^2} + \frac{2\sqrt{2\pi}}{\gamma \tau' \Delta\omega T_2}}.$$

Figure IX-10 shows the experiments of Henry and Bordé, and a theoretical curve fitted to the curve of γ vs pressure. To fit the last curve, we assumed that Γ , $1/\tau$, and



Fig. IX-10. Bordé and Henry's experimental results. Circles are derived from our formula.

 $1/T_2$ are all proportional to pressure, a reasonable assumption for CO_2 . Furthermore, we adjusted the farthest point to fall on the center of the error bar. This was done by assuming that $\gamma \tau' = 3$.

It is clear that the curve thus obtained is a better fit than the straight line plotted by Bordé and Henry.⁵

H. A. Haus, P. W. Hoff

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(IX. ELECTRODYNAMICS OF MEDIA)

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