Transients in a far-infrared laser

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The transient behavior of an optically pumped, monomode, far-infrared laser is investigated with the use of on-off switching of the pump power. Various regimes of the onset of laser action are observed and described with the use of a two-level model for the lasing medium. At low pressures there is a possibility of undamped oscillation for the output power, which is assigned to a competition between two fields. The transients observed as the pump power is switched off are explained with the use of a three-level model.

I. INTRODUCTION

The transient behavior of a continuous wave far-infrared (far-ir) laser has not been investigated so far at a submillisecond time scale. However, it gives rise to a class of phenomena of interest according to different viewpoints. The most interesting situation is obtained by switching the pump laser radiation which creates the population inversion responsible for the far-ir laser action. The output power of the laser is monitored as laser radiation builds up or stops. As we investigate here the behavior of a cw laser, the gain in the active medium remains relatively low and the time scale of the transients observed is in the 0.1- to 10-μs range.

The buildup of radiation in a cw laser was considered theoretically by Lamb in the low-saturation regime.[1] Using a perturbation expansion of the gain coefficient, he showed that laser action builds up in an aperiodic way with a delay time and a rise time decreasing as the gain increases. His calculation was soon confirmed by an experiment of Pariser and Marshall who obtained very good agreement between Lamb's calculation and the corresponding experimental results observed on a cw He-Ne laser which is Q switched.[2] About ten years later, the same technique was used by Arecchi and De Giorgio, who investigated the statistical properties of laser radiation during the transient regime.[3] Because of experimental limits, these two works were restricted to low-saturation conditions. On the other hand, the far-ir laser allows one to explore the whole range of saturation parameters starting from just above threshold to highly saturating fields. The present paper is devoted to average signals which result from the sum over a large number of experiments (typically 2^{12}). The statistical properties of these signals will be discussed in a forthcoming paper.[4]

The experimental results reported here are of interest in connection with three kinds of studies. First, the far-ir laser exhibits a class of transients which is qualitatively different from the coherent transients (optical nutation, optical precession, photon echoes, . . .) observed when the interaction of molecules or atoms with a cw radiation field is suddenly switched by, e.g., a Stark field shifting the absorption frequencies.[5,6] Secondly, it is an example of cooperative behavior of a quantum system such as superradiance[7] or lethargic gain,[8] but in our experiments the system is continuously pumped so its dynamics are changed by the continuous coupling with the pump radiation. Note also that here the system is enclosed in a cavity while in most superradiance experiments the sample is in a single pass cell, although there has been recent experiments on superradiance of Rydberg atoms in a cavity.[7(b)] Thirdly, the cw far-ir laser in the transient regime is a simple example of a system passing from an initially unstable to a finally stable state, a problem which is of interest for people concerned with synergetics.[9] There the far-ir laser is particularly noteworthy since, in most cases, it is described by a simple theoretical model: the homogeneously broadened two-level laser. The comparison with predictions from this model is, of course, invaluable in understanding the various effects occurring near the onset of laser radiation.

The limits and advantages of the experiments come from the choice of switching a cw pump. Using a TEA (transverse excitation atmospheric) CO_2 laser would give rise to transients in the nanosecond region but the pulse shape of this laser makes difficult a simple analysis of the dynamics of the far-ir laser in that case.[10] Square wave switching of the cw pump power allows clean demonstration of the phenomena at the price of a restricted time scale. In particular, it is impossible to observe effects faster than the rotational relaxation, such as superradiance. On the other hand, those phenomena occurring in the millisecond range, such as the decrease of laser efficiency due to vibrational bottleneck,[11] will be neglected here.

A preliminary report of some of the results discussed here was given in Ref. 12. The onset of far-ir laser action in the low-gain region was described both theoretically and experimentally. These results have been extended to the high-gain region. Section II summarizes the corresponding experimental data while the theoretical model is given in Sec. III together with numerical solutions. An undamped oscillating regime was observed in far-ir lasers[13] and attributed by Lawandy to the Statz—de Mars instability.[14] On the other hand, the undamped oscillation of the output power of our far-ir laser has been attributed to another phenomenon which is discussed in Sec. IV.

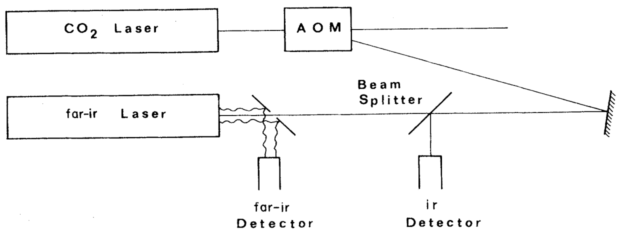

FIG. 1. Experimental setup.

Transients occurring as the laser stops may not be explained using the two-level model since, in that case, the pumping and emission processes are in the transient regime simultaneously. The experimental results and the corresponding calculations are given in Sec. V. As shown in Sec. II, the laser needs some incubation time before its output power builds up rapidly. The corresponding time delay depends not only on the linear gain but also on the intensity present inside the cavity at the beginning of the process. This intensity may be varied, using what is left-over from a preceding pulse experiment and varying the interruption time between the two excitation pulses. The results of such experiments are given and discussed in Sec. VI.

II. EXPERIMENT

A. Experimental setup

All the experiments reported here were made using a monomode Fabry-Pérot far-infrared laser which was previously described in detail.[15] Similar experiments were also carried out in a metallic waveguide laser and gave similar results. However, the field distribution is better known in the Fabry-Pérot laser and there is much less mode overlapping. So, for the sake of simplicity, only those results obtained with the Fabry-Pérot laser are reported. All except those of Fig. 2 are related to signals observed from the 743-μm line of HCOOH although an identical behavior was observed on the other lasing lines which were investigated. This particular line was chosen because it gives reasonably strong far-infrared laser action and because its relatively low frequency allows us to use a point-contact W-Si ("cat-whisker") detector.

The experimental setup is schematically shown on Fig. 1. The output power from a cw CO_2 laser oscillating on the 9-μm R (40) CO_2 line is limited to 3 W to avoid thermal runaway of the acousto-optic modulator which switches the laser radiation with a rise time less than 100 ns. The 1-W infrared beam coming out of the modulator is monitored by a fast HgCdTe detector which gives the time reference for the delay measurements and it is simultaneously sent into the far-ir laser cavity. The far-ir emission is monitored by the cat-whisker detector which, in addition to a suitable preamplifier, provides a detection bandwidth up to 20 MHz. The signal is then fed into a digital averager and possibly into a microcomputer for further data processing.

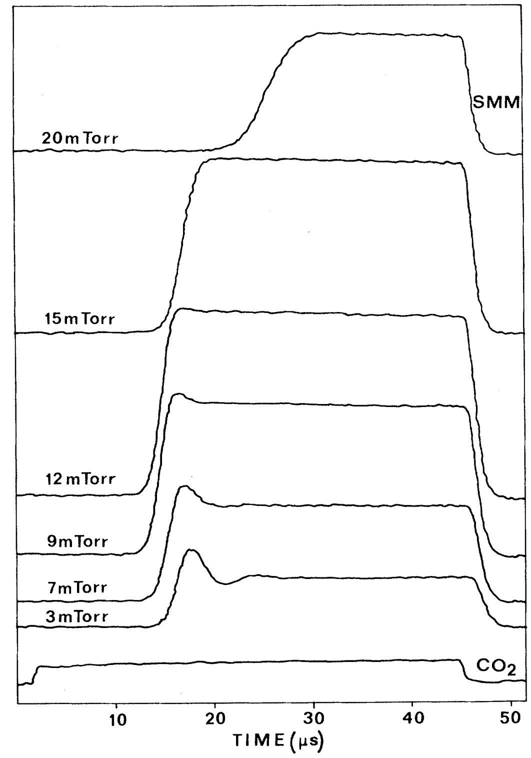

FIG. 2. Pressure dependence of the far-ir laser transients at the onset of laser power. ir and far-ir lasers are tuned to resonance. ir power: 0.8 W. Far-ir emission line: H^{13} COOH at 788 μm. ir pump 9P12 of CO_2 laser.

B. Onset of far-ir laser action

As the pump power is switched on, the laser output power does not set up immediately, but stays below any detectable value for a relatively long time, of the order of tens of microseconds, until it eventually jumps to its steady-state value in a time of the order of a few microseconds. This is illustrated in Fig. 2 where the buildup of laser emission is displayed for various pressures. Similar results were obtained by varying the pump power or the far-ir laser tuning. They all show that two regimes may be observed: (i) a low-saturation regime where the laser output sets up aperiodically and (ii) a regime in which the steady-state level is reached through damped oscillations. The aperiodic regime is observed in low-power—high-pressure situations when the active medium is only weakly saturated while the oscillations require strong saturation. This behavior is somehow similar to pure Rabi nutation where an absorbing gas is suddenly brought into resonance with a cw field. The transmitted power then exhibits damped oscillations at the frequency $\mu E/\hbar$, where μ is the transition electric dipole moment and E the amplitude of the cw field. For these oscillations to be observable, the Rabi frequency must be larger than the unsaturated linewidth γ, thus giving the saturation condition $\mu E/\hbar\gamma > 1$. In the case of the laser oscillation buildup, the driving field E is not constant since it is the actual field inside the laser which drives its own "nutation." Note that a similar situation is encountered in self-induced transparency and in superradiance where, because of the shorter time scale involved, additional propagation effects have to be considered.

The rise of far-ir laser emission in the aperiodic regime was previously reported in Ref. 12 where it was shown that this regime could be very satisfactorily described by Lamb's semiclassical theory of the laser.[1] Using a perturbation expansion of the polarizability in successive powers of the electric field and considering the polarization as a source term, the equation of motion for the laser intensity is obtained:

$$\dot{I}=2I(\alpha-\beta I)\ , \tag{1}$$

where α is the linear net gain coefficient and β is the self-saturation coefficient. This equation may be analytically solved:

$$I=\frac{\alpha[I_0/(\alpha-\beta I_0)]\exp(2\alpha t)}{1+\beta[I_0/(\alpha-\beta I_0)]\exp(2\alpha t)}\ , \tag{2}$$

where $I_0=I(0)$ is the intensity present in the laser cavity at time $t=0$ when laser action starts. An excellent agreement between this calculated result and experimental data was obtained as long as the active medium was not heavily saturated. In the saturated medium, the perturbation expansion of the polarizability is no longer a good approximation. Moreover, for obtaining Eq. (2) it was assumed that the electric field did not vary much during a time of the order of γ^{-1}. As the rise time of laser power is of the order of α^{-1}, this condition is not fulfilled in saturation conditions. Thus to describe the buildup of laser power in the strong-saturation regime a new model is required as explained in Sec. III.

III. BUILDUP OF OSCILLATION IN THE STRONG-SATURATION SITUATION

A. Model

The exact solution of the laser equations requires integrations over, for instance, the transverse and longitudinal field variations. The aim of this subsection is to derive simple equations for the laser, making reasonable assumptions so that the main features of the laser transients may be derived from these equations without carrying all the lengthy integrations.

A three-level model is usually needed to describe the various processes involved in the optically pumped far-ir lasers. In most experiments it is, however, a reasonable assumption to neglect the influence of the infrared transients and thus to use a two-level model with a pumping rate. The following reasons support this model: (i) The delay for rise of far-ir output power is of the order of 10 μs or more. In usual operating conditions, all infrared transients are damped out since the delay is much longer than the rotational relaxation time. (ii) The damping of ir transients is even faster when Doppler broadening and ir field inhomogeneities are considered. Although it describes a gas laser, the model described here may neglect the inhomogeneous broadening of the far-infrared transition. In fact, the population inversion is created by optical pumping with an infrared laser. Because only one velocity group is in resonance with the pump radiation there is a velocity selection through optical pumping and only a very narrow velocity group is transferred to the excited vibrational state. Moreover, if the ir radiation is exactly on resonance with the ir transition, the particular velocity group which is pumped is that with a zero component along the field direction so there is no Doppler shift. In very special conditions, Doppler broadening and shift are responsible for new features which are described in Sec. IV.

When conditions (i) and (ii) are not fulfilled a three-level model like the one described in Sec. V is required. Using both two- and three-level models, it is easy to separate what comes out of single- far-ir photon and two- (ir + far-ir) photon processes. Common assumptions are needed to set up these models. It is therefore useful to give here, in some detail, the procedure used in the derivation of the basic equations.

Following Lamb's method, the far-ir field inside the cavity is developed on the modes of the passive cavity:

$$E_s(z,t)=\sum_n E_{s,n}(t)\sin(k_n z)\ , \tag{3}$$

with $k_n=\Omega_n/c$, Ω_n is the resonance frequency of mode n. As in our laser, the mode spacing is larger than the mode width

$$E_{s,n}(t)=E_{s,n}(0)\cos[\omega_n t+\varphi_n(t)]\ .$$

The transverse variation of the electric field is neglected since the laser is operating in high-symmetry modes.[16] As the field is linearly polarized, its vector nature is omitted.

The Fabry-Pérot resonator has been chosen because its various modes are well separated so that the far-ir laser is operating monomode. Then only one term is retained in the sum of Eq. (3).

The polarization $P_s(z,t)$ induced by this field is also developed on the modes n:

$$P_s(z,t)=\sum_n P_{s,n}(t)\sin(k_n z)$$

with

$$P_{s,n}(t)=P_{s,n}(0)\cos[\omega_n t+\varphi_n(t)]\ .$$

P_s and E_s refer to quantities in which the fast variation $(\sim\omega_n^{-1})$ is included.

The polarization and electric field are coupled by Maxwell equations which are written, in this case, as

$$-\frac{\partial^2 E_s(z,t)}{\partial z^2}+\mu_0\sigma\frac{\partial E_s(z,t)}{\partial t}+\mu_0\epsilon_0\frac{\partial^2 E_s(z,t)}{\partial t^2}=-\mu_0\frac{\partial^2 P_s(z,t)}{\partial t^2}\ .$$

For the mode n the conductivity σ_n is related to the quality factor Q_n by $\sigma_n=\epsilon_0\omega_n/Q_n$, where ω_n is the frequency of the laser operating on mode n.

Because of the nonlinearity of the coupling between P_s and E_s there may exist in the development of P_s spatial harmonics different from $\sin k_n z$ even in monomode operation of the laser. Separating the real $C_{s,n}(t)$ and imaginary $S_{s,n}(t)$ parts of the projection $P_{s,n}(t)$ of the polarization on mode n

$$\begin{aligned}&\dot{E}_{s,n}(t)+\frac{\omega_n}{2Q_n}E_{s,n}(t)=\frac{1}{2}\frac{\omega_n}{\epsilon_0}S_{s,n}(t) ,\\&[\Omega_n-\omega_n-\dot{\varphi}_n(t)]E_{s,n}(t)=\frac{1}{2}\frac{\omega_n}{\epsilon_0}C_{s,n}(t) .\end{aligned} \tag{4}$$

The first Eq. (4) relates the field derivative to the gain in the medium and the cavity losses. The second one shows that the laser frequency may be different from that of the passive cavity because of dispersion in the gain medium. Note that $C_{s,n}$ and $S_{s,n}$ are components of the *global* polarization

$$P_{s,n}(t)=(2/L)\int_0^L P_s(z,t)\sin(k_n z)dz$$

for a cavity length L while density-matrix elements ρ_{ij} give the *local* polarization

$$P_s(z,t)=\mu_r\int_0^t \lambda(z,t_0)[\rho_{23}(z,t,t_0)+\rho_{32}(z,t,t_0)]dt_0 ,$$

where $\lambda(z,t_0)$ is the pumping rate of active molecules at time t_0, μ_r is the matrix element of the electric dipole moment between levels 2 and 3 connected by the far-ir laser transition, and the pump is switched on at time $t=0$. The local polarization is easily expressed in terms of density matrix elements averaged over t_0

$$P_s(z,t)=\mu_r\rho_{23}(z,t)+\text{c.c.}$$

The evolution equation of the density matrix in the rotating frame may be expressed as

$$\begin{aligned}&\dot{N}(z,t)=-\gamma[N(z,t)-N^0(z,t)]-\frac{E_{s,n}(t)}{\hbar}S_s(z,t)\sin(k_n z) ,\\&\dot{C}_s(z,t)=-\gamma C_s(z,t)-[\omega_{23}-\omega_n-\dot{\varphi}_n(t)]S_s(z,t) ,\\&\dot{S}_s(z,t)=-\gamma S_s(z,t)+[\omega_{23}-\omega_n-\dot{\varphi}_n(t)]C_s(z,t)\\&\qquad+\frac{\mu_r^2E_{s,n}(t)}{\hbar}N(z,t)\sin(k_n z) ,\end{aligned}$$

where $N(z,t)=\rho_{22}(z,t)-\rho_{33}(z,t)$; $C_s(z,t)$ and $S_s(z,t)$ are, respectively, the imaginary and real parts of $P_s(z,t)$ and N^0 is the population inversion in the absence of far-ir laser action $N^0=\lambda(z,t)/\lambda$. Introducing dimensionless quantities

$$\begin{aligned}&\xi=\frac{2Q_n\gamma}{\omega_n} ,\quad \eta=\frac{\mu_r^2\bar{N}Q_n}{\hbar\epsilon_0\gamma} ,\quad \Delta_1=2Q_n\frac{\Omega_n-\omega_n}{\omega_n} ,\\&\Delta_2=\frac{\omega_{23}-\omega_n}{\gamma} ,\quad u=\frac{N(z,t)}{\bar{N}} ,\quad u^0=\frac{N^0(z,t)}{\bar{N}} ,\\&y_1=\frac{C_s(z,t)}{\bar{N}\mu_r} ,\quad Y_1=\frac{C_{s,n}(t)}{\bar{N}\mu_r} ,\quad y_2=\frac{S_s(z,t)}{\bar{N}\mu_r} ,\\&Y_2=\frac{S_{s,s}(t)}{\bar{N}\mu_r} ,\quad X_s=\frac{\mu_rE_{s,n}(t)}{\hbar\gamma} ,\\&\bar{N}=\frac{1}{L}\int_0^L N^0(z,t)dz ,\quad Y_i=\frac{2}{L}\int_0^L y_i\sin(k_n z)dz ,\end{aligned} \tag{5}$$

where ξ is the relaxation rate in reduced inverse time units, u is proportional to the population inversion in the zero far-ir field, and η characterizes the unsaturated far-ir gain. The components of the polarization are given in units of $\bar{N}\mu_r$, the far-ir Rabi frequency X_s, and detuning Δ_2 in units of the homogeneous linewidth γ. Bloch-Maxwell equations may be written as

$$\begin{aligned}&\dot{X}_s=-X_s+\eta Y_2 ,\\&\dot{\varphi}_n=\Delta_1-\eta(Y_1/X_s) ,\\&\dot{u}=-\xi[u-u^0+X_sy_2\sin(k_n z)] ,\\&\dot{y}_1=-\xi(y_1+\Delta_2y_2)+\dot{\varphi}_ny_2 ,\\&\dot{y}_2=-\xi[y_2-\Delta_2y_1-uX_s\sin(k_n z)]-\dot{\varphi}_ny_1 ,\end{aligned} \tag{6}$$

where the dot means a time derivative with respect to the reduced time $t'=\omega_n t/2Q$ which gives a time scale well fitted to the problem, i.e., in units of the photon lifetime in the cavity. Capital letters Y_1, Y_2, and X_s refer to global quantities while lower case letters u, u^0, y_1, and y_2 indicate local quantities, which are related by equations similar to (5).

The gain coefficient η is easily calculated as a function of the ir field $E_p(z,t)$ using a two-level model for the ir transition. In the Doppler limit,

$$\eta=\frac{N_1^0\mu_r^2Q_n}{2\hbar\epsilon_0\gamma}\frac{\epsilon_p\Pi^{1/2}I_p}{(1+I_p)^{1/2}} ,$$

where I_p is the reduced infrared intensity: $I_p=(\mu_v^2E_p^2/2\hbar^2\gamma^2)$, ϵ_p is proportional to the ratio of the homogeneous $\Delta\nu_H$ and Doppler $\Delta\nu_D$ widths: $\epsilon_p=(\ln 2)^{1/2}(\Delta\nu_H/\Delta\nu_D)\ll 1$, γ is the rotational relaxation time and N_1^0 is the population of level 1 at thermal equilibrium.

When far-ir and ir fields are exactly on resonance ($\Delta_1=\Delta_2=0$) the dispersive terms are decoupled from the other ones and the system of five equations is separated into three equations for X_s, u, and y_2 and two equations characterizing terms which are not observable in our experiments

$$\dot{X}_s=-X_s+\eta y_2 , \tag{7a}$$

$$\dot{u}=-\xi[u-u^0+X_sy_2\sin(k_n z)] , \tag{7b}$$

$$\dot{y}_2=-\xi[y_2-uX_s\sin(k_n z)] . \tag{7c}$$

B. Limit cases $\xi\ll 1$ and $\xi\gg 1$

The relative evolution rate of field and molecular variables depends mainly on the parameter ξ. In our experiments, this parameter may span a large range of values and it is useful to consider two limit cases.

If the lifetime of the photons inside the cavity is larger than the inverse relaxation rate ($2Q_n/\omega_n\gg\gamma^{-1}$), ξ is much larger than unity. Then the rise time of the output power which is always limited by $2Q_n/\omega_n$ is longer than rotational relaxation time and the field varies little during γ^{-1}. This is a validity condition of Lamb's perturbation treatment. In this situation, population inversion and polarization are "fast" variables which are always in equilibrium with the field and molecular variables may be adiabatically eliminated $\dot{u}=\dot{y}_1=\dot{y}_2=0$. Then

$$y_2=u^0\frac{(1+\Delta_2^2)^{-1}X_s\sin(k_nz)}{1+X_s^2\sin^2(k_nz)(1+\Delta_2^2)^{-1}}\ , \tag{8}$$

where second-order terms have been neglected.

The global quantity is obtained by (5), assuming that u_0, the population inversion at zero field, varies little over one wavelength, i.e.,

$$\frac{2}{L}\int_0^L u^0\sin^2(k_nz)dz=\frac{1}{L}\left[\int_0^L u^0dz-\sum_{j=0}^{n-1}u^0\left[j\frac{\lambda}{2}\right]\int_{j(\lambda/2)}^{(j+1)(\lambda/2)}\cos(2k_nz)dz\right]=1\ .$$

Then one obtains

$$Y_2=\frac{X_s}{1+\Delta_2^2}-\frac{3}{4}\frac{X_s^3}{(1+\Delta_2^2)^2}$$

and

$$\dot{X}_s=X_s\left[\frac{\eta}{1+\Delta_2^2}-1-\tfrac{3}{4}\eta\frac{X_s^2}{(1+\Delta_2^2)^2}\right]\ ,$$

which is exactly Lamb's result for the aperiodic regime.

At low pressures $\xi \ll 1$, molecular variables are "slow" relative to the electric field which is the fast variable. The electric field then follows the molecular variables and $X_s=\eta Y_2$. This situation corresponds to the saturation damped oscillation regime where the laser output exhibits damped oscillations before reaching its steady state. Its behavior is analogous to coherent transients in the microwave region, when the response of a system subjected to a fast change of the excitation field shows damped Rabi nutation[17(a)] or Stimulated Inelastic Resonance Fluorescence (SIRF) [Ref. 17(b)]. However, in the case of the laser, the interpretation is made more difficult because the far-ir field and the Rabi frequency are varying simultaneously. In that case, the passage from local to global variables has to be examined specifically.

C. Passage from local to global variables

The calculation of global variables from (7b) is easy but the main difficulty arises from (7c) since the product term introduces coupling of spatial harmonics of different orders. $u(z,t)$ is easily shown to be developable over even terms $\cos 2jk_nz$. Similarly, $y_2(z,t)$ may be developed as a sum of odd order terms $\sin(2j+1)k_nz$. Generally speaking, every term of order l is coupled to terms of order $(l\pm1)$ and there is an infinite set of coupled differential equations to be solved.

The corresponding steady-state problem has been solved by Stenholm and Lamb using a continuous fraction expansion.[18] Here we make the rate equation approximation in which the spatial harmonics development of u and y is restricted to the lowest-order term

$$u(z,t)=u_0(t)=U\ ,$$

$$y_2(z,t)=Y_2(t)\sin(k_nz)\ .$$

Then

$$\begin{aligned}\dot{X}_s&=-X_s+\eta Y_2\ ,\\ \dot{U}&=-\xi(U-1+\tfrac{1}{2}X_sY_2)\ ,\\ \dot{Y}_2&=-\xi(Y_2-UX_s)\ .\end{aligned} \tag{9}$$

In the following, we consider that both radiations are exactly resonant ($\Delta_1=\Delta_2=0$).

This nonlinear system of differential equations is similar to those obtained by Graszyuk and Orayevskiy in their investigation of transient processes in masers[19] and by Haken in his semiclassical theory of the laser.[20] It should be noticed that all the validity conditions for the rate-equation approximation are not fulfilled here. In particular, there is a rapid variation of the population inversion along the cavity axis. While, for lasers working in the visible range, one can assume that an atom travels along many wavelengths during an average interaction time, i.e., its average velocity is such that $v/\gamma \gg \lambda$, this is no longer valid for the long wavelengths at which the far-ir laser is operating.

Although the global problem may be solved on a computer, it is useful to derive such a set of equations since some general properties of the solutions may be derived from it without knowing the exact form of the solutions.[21,22] They will be used for a linear stability and bifurcation analysis[23] of the instability appearing when $\eta > \xi^{-1}(1+4\xi)/(1-2\xi)$, which will be published in a following paper.

D. Numerical solutions

Equations (9) are easily solved numerically. Figure 3 gives examples of calculated transients obtained for various pressures inside the far-ir laser cavity. Values chosen for obtaining the waveform of Fig. 3 are typical values for the far-ir lasers.[11,12] They give limit pressures for the oscillation of the laser very close to those experimentally observed with our setup since it is oscillating from 2 to 35 mTorr when pumped with 1-W CO_2 laser radiation.

The variation range for the time delay T_D and rise time T_r are in good agreement with experimental results. The large values of T_D and T_R obtained at high pressure are characteristic of the low-saturation regime. As the pressure decreases calculated waveforms exhibit an overshot particularly visible at low pressures. Experimentally, this overshooting is more damped because of inhomogeneity of the ir pumping, transverse dependence of the far-ir mode,[17(a)] etc. Nevertheless, calculated shapes using the two-level model give a very good description of transients observed at the onset of the far-ir laser field, even when this field is strongly saturating.

IV. RELAXATION OSCILLATIONS IN LASERS

It has long been observed that under cw excitation conditions some single-mode lasers might deliver a pulsed output even if there is no saturable absorber inside the

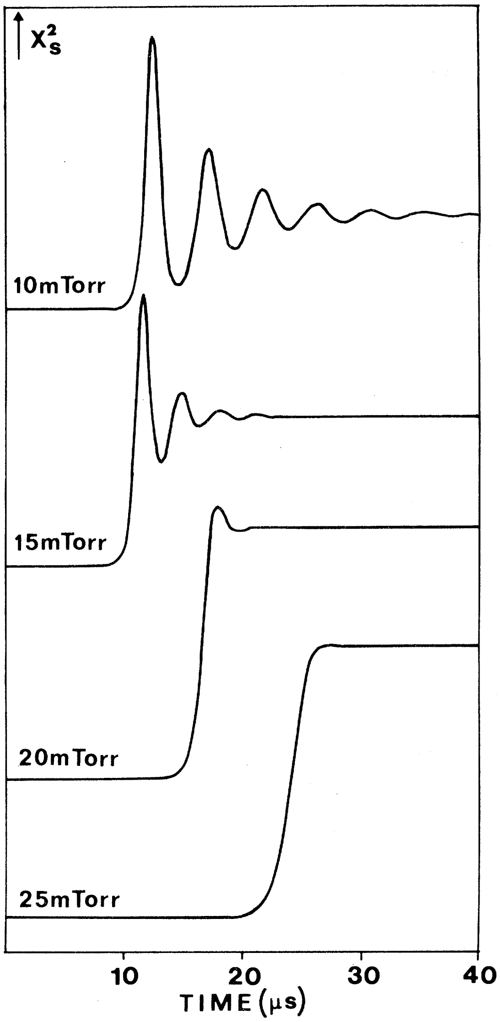

FIG. 3. Pressure dependence of the far-ir laser transients at the onset of laser power as calculated using (9) ir intensity 8.10^{-2}-W cm^{-2} far-ir cavity quality factor $Q=5.10^5$. Doppler width 0.9 kHz/GHz. Collision width = 20 MHz Torr^{-1}. Partition function 8800. Far-ir dipole moment $\mu_r=0.7$ D. ir electric dipole moment $\mu_v=0.05$ D. In all curves peak intensities are normalized to unity.

laser cavity. There has recently been a renewal of interest in this field in connection with synergetics. If the onset of laser action is regarded as a first-order phase transition like behavior, there could be some other phases and possibly unstable states. The first investigation of the relaxation oscillations by Statz and De Mars used a rate equation approach[14] but it was later shown by Makhov[24] and Sinnett[25] that their equations cannot give rise to an instability. Risken and Nummedal made a stability analysis of the system of partial differential equations that they set up to describe the laser without the spatial Fourier development.[26] They showed that a cw solution cannot exist for a certain choice of time-independent laser parameters.[4] Using a different approach, Casperson then demonstrated that the single-mode solution found initially in a laser is unstable if additional sidebands exist and if the laser field is saturating enough to induce dispersion effects which allow the sideband frequencies to have the same wavelength as the saturating field.[27]

These kind of mode splitting instabilities, for which other frequencies having the same mode structure as the initial oscillating field have positive gain, was recently demonstrated by Maeda and Abraham[28] on an inhomogeneously broadened 3.5-μm Xe laser. They also showed that other (subharmonic) bifurcations occurred as the suitable parameter was further increased and lead to a route from stability to chaos in a single-mode laser.

In the far-ir region undamped oscillations were already reported by Lawandy and Koepf[13] and Vass *et al.*[29] However, the conditions required for these oscillations to appear are not well established. Moreover, they were misinterpreted using the Statz-De Mars rate equations in which the output always eventually damps back to the steady-state value. So there is a need of reinvestigating the relaxation oscillations in the far-ir laser in order to get information on their dependence on experimental parameters such as far-ir laser frequency, ir laser power, and ir frequency.

In our Fabry-Pérot laser, relaxation oscillations were observed only at pressures below 6 mTorr. They disappeared when the ir radiation was tuned to achieve the maximum far-ir output power; in fact, they required an ir frequency offset of about 10 MHz or more. However, the frequency of these relaxation oscillations depends little on the ir laser frequency. On the other hand, the phenomenon depends strongly on the far-ir cavity tuning: the undamped oscillations were observed only when the far-ir cavity length was set to the center of the emission mode while they became quickly damped when the cavity was slightly (~ 0.5 μm) detuned from resonance as shown on Fig. 4. Our interpretation of the undamped oscillations is based on the fact that they are observed *if and only if* the far-ir mode (i.e., the dependence of the far-ir power on far-ir cavity length) presents the two-peak shape already observed by other groups[30,31] and explained using rate equations by De Temple and Danielewicz.[32]

At low pressures, the far-ir transition is Doppler broadened. In this situation, the velocity groups pumped by an off resonance ir radiation have positive gain for different frequencies. More precisely, if the pump laser is shifted off resonance by an amount $\Delta\omega_{\mathrm{ir}}$, the velocity groups which are pumped by the direct and reflected ir beams are such that

$$\frac{v}{c}=\pm\frac{\Delta\omega_{\mathrm{ir}}}{\omega_{\mathrm{ir}}}$$

and the gain profile will be peaked around values such as

$$\omega_{\text{far-ir}}=\omega^{0}_{\text{far-ir}}\left[1\pm\frac{v}{c}\right]=\omega^{0}_{\text{far-ir}}\left[1\pm\frac{\Delta\omega_{\mathrm{ir}}}{\omega_{\mathrm{ir}}}\right],$$

where the first $\pm$ sign comes from the standing wave far-ir field. When the far-ir Doppler shift $\omega_{\text{far-ir}}(v/c)$ is larger than the homogeneous width, the far-ir gain curve is two-peaked. The frequency difference between these two

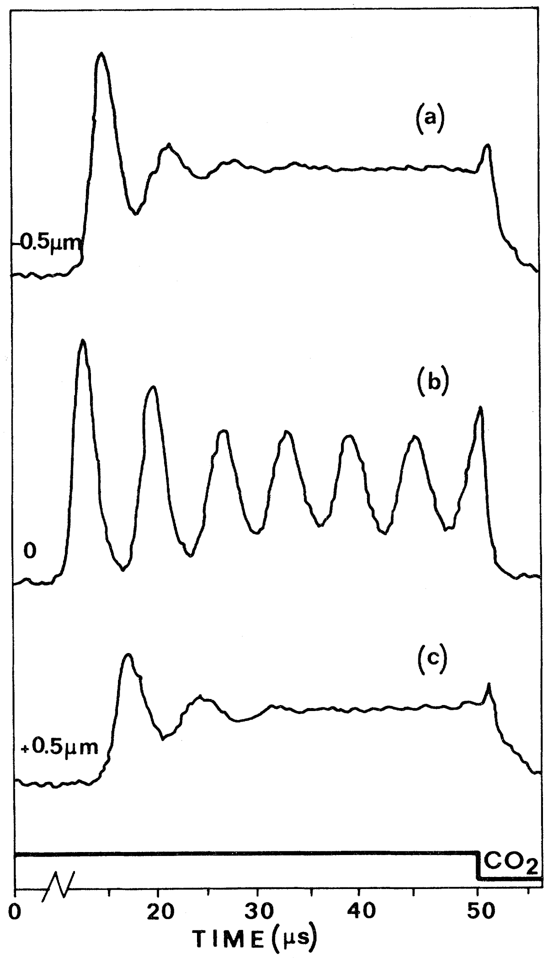

FIG. 4. An example of transients on the 742-μm line of H^{12} COOH due to mode splitting for the far-ir cavity on resonance (b) and slightly off resonance [(a) and (c)]. For (a) and (c) the cavity length is changed by 0.5 μm. HCOOH pressure: 4 mTorr. ir pump offset ~ 10 MHz. ir power: 830 mW.

peaks on the far-ir mode (typically 500 kHz) is consistent with the ir offset (typically 10 MHz).

This shows that the two-peak spectrum is associated with a single class of molecules in the same state and in the same velocity group. From our experimental results it clearly comes out that the observation of the instability requires (i) a two-peak spectrum for a single class of molecules and (ii) a centrally tuned cavity as illustrated in Fig. 4.

The undamped oscillations originate from the competition between two oscillating fields which may be explained as follows: when the two-peak curve is observed and the far-ir cavity is tuned halfway between the peaks, two fields with frequency equally shifted from the cavity frequency may exist simultaneously due to the frequency pushing and pulling terms in the laser frequency determining Eq. (4). Because of Doppler effect the gain in each group is shared between the two oscillation fields and there is a possibility of competition between these fields. The amount of competition depends on the overlapping between the two components of the far-ir mode curve. This quantity may be varied by adjusting the pump frequency offset which sets the frequency difference between the maximums of the two peaks. The simultaneous existence of different resonance frequencies in a single-mode laser was convincingly demonstrated by Casperson[27] both in an homogeneously and in an inhomogeneously broadened laser using graphical solution of resonance conditions for highly dispersive media.

Instabilities appearing in lasers with a two-peaked spectrum and a centrally tuned cavity were theoretically considered by a number of authors, either in the case of the Lamb dip, of holes burned in an inhomogeneously broadened gain profile,[27,33,34] or in lasers with saturable absorbers,[35] who showed that in such cases there is a possibility of high-frequency oscillations in the laser output.

An over simplified model of the instability is provided by the coupled equations for a two-mode laser as given by Lamb.[1] Although they are only approximations, they give a sufficient picture for a qualitative fit of theory to our experiment. Let I_1 and I_2 be the intensities of the two fields. The gain equations for these quantities are

$$\begin{aligned} \dot{I}_1 &= 2(\alpha_1 - \sigma_{12} I_2) I_1 - 2\beta_1 I_1^2 , \\ \dot{I}_2 &= 2(\alpha_2 - \sigma_{21} I_1) I_2 - 2\beta_2 I_2^2 , \end{aligned} \tag{10}$$

where $\alpha_1(\alpha_2)$ and $\beta_1(\beta_2)$ represent, respectively, the linear and self-saturation coefficients for $I_1(I_2)$. σ_{12} and σ_{21} are cross saturation terms which describe the decrease of the gain for the field I_1 due to the presence of I_2 and vice versa. Experimentally, the σ_{12} and σ_{21} terms may be varied by detuning the ir laser.

Lamb's results of stability analysis of Eqs. (10) may be adapted to our situation. They show that:

If the far-ir cavity is slightly detuned and favors one of the peaks, the corresponding field will dominate and the cross saturation term will decrease the gain on the other field.

In the strong coupling limit ($\sigma_{12}\sigma_{21}/\beta_1\beta_2 > 1$) two frequencies cannot oscillate simultaneously. Depending on the initial conditions one "mode" or the other is favored and the laser exhibits a bistable behavior, then there is no oscillation on the output.

If the net gain on each mode is positive and in the weak coupling case ($\sigma_{12}\sigma_{21}/\beta_1\beta_2 < 1$) the two fields may exist at the same time. They are observed as a beating on the far-ir detector. Experimentally, this is observed when the far-ir cavity is tuned midway between the two modes to avoid favoring of one mode and if the ir laser is reasonably detuned to prevent strong coupling. This is exactly the situation in which the relaxation oscillations were observed.

V. LASER FALL TRANSIENTS

The two-level model which was adequate for transients observed as laser radiation builds up may not be used when the pump is switched off. In that case the pumping and the emission transients have to be considered simultaneously. This requires a three-level model which is

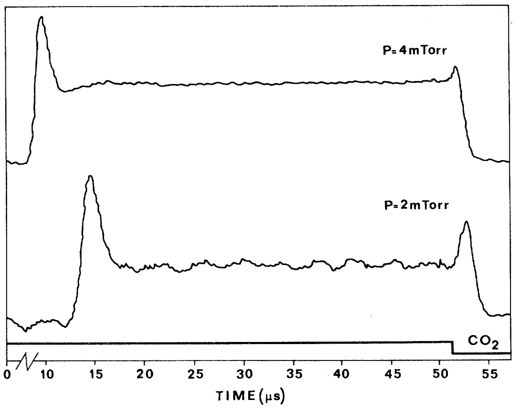

FIG. 5. Experimental results of far-ir laser transients at the off-switching of the pump radiation ir power: 830 mW. HCOOH pressure: 2 and 4 mTorr.

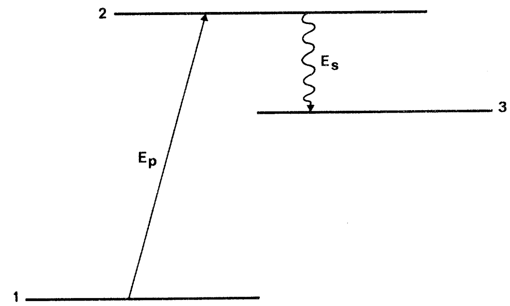

FIG. 6. Three-level scheme for the far-ir laser.

described here.

Before going into the calculations, let us first give the basic experimental results. Examples of observed signals are given in Fig. 5, where it is shown that as the CO_2 radiation is switched off the output power does not always decrease. A decrease is expected because of the collisional damping of the polarization and the finite lifetime of the photons in the cavity. Attempts to fit the decreasing part of the signals to single exponentials were unsuccessful. In fact, when the far-ir medium is strongly saturated, i.e., at pressures lower than 7 mTorr, an increase of the output power is observed as the pumping is switched off. This overshot is due to the stop of the destructive interference between single and two-photon processes which lowers far-ir laser efficiency when the pump is on. In absence of pump radiation the two-photon process cancels and there is no more destructive interference. Similar effects were observed in pump switched three-level paramagnetic masers by Manenkov *et al.*[36] and in microwave-microwave double resonance by Glorieux and Macke.[37] In the latter experiments the situation is somehow different since absorption transients were detected.

In the strong collision approximation, neglecting the spatial dependence and using the same simplifications as in Sec. III C, the following equations may be derived from the density matrix equations of a three-level system subjected to two resonant electromagnetic fields as shown in Fig. 6:

$$\begin{aligned}
\dot{U} &= -\xi(U + X_s Y_s - \tfrac{1}{2} X_p Y_p)\ , \\
\dot{W} &= -\xi(W - W^0 - X_p Y_p + \tfrac{1}{2} X_s Y_s)\ , \\
\dot{Y}_p &= -\xi(Y_p - \tfrac{1}{2} X_s Y_{sp} + X_p W)\ , \\
\dot{Y}_s &= -\xi(Y_s - X_s U + \tfrac{1}{2} X_p Y_{sp})\ , \\
\dot{Y}_{sp} &= -\xi(Y_{sp} + \tfrac{1}{2} X_s Y_p - \tfrac{1}{2} X_p Y_s)\ ,
\end{aligned} \tag{11}$$

where we have used dimensionless quantities analogous to those of Sec. III. We now have the following definitions:

$$U = \frac{(\rho_{22} - \rho_{33})}{N^0}\ , \quad W = \frac{(\rho_{22} - \rho_{11})}{N^0}\ , \quad Y_s = \frac{i(\tilde{\rho}_{23} - \tilde{\rho}_{32})}{N^0}\ ,$$

$$Y_p = \frac{i(\tilde{\rho}_{12} - \tilde{\rho}_{21})}{N^0}\ , \quad Y_{sp} = \frac{(\tilde{\rho}_{13} + \tilde{\rho}_{31})}{N^0}\ ,$$

$$W^0 = \frac{(\rho_{22}^0 - \rho_{11}^0)}{N^0}\ ,$$

with $\tilde{\rho}_{ij} = \rho_{ij}\exp(-i\omega_{ij}t)$, where ω_{ij} is the angular frequency of the $i \to j$ transition. They are all given in units of N^0, U (W) is the population difference of levels connected by the far-ir (ir) field, Y_s and Y_p characterize the far-ir gain in the medium and the absorption of the ir field, Y_{sp} is related to the coherence between levels 1 and 3, and W^0 is the value of W at thermal equilibrium.

The coupling between the far-ir polarization and the far-ir field is given by the same equation as in the two-level system:

$$\dot{X}_s = -X_s + \eta Y_s\ .$$

The overshot observed as the pump is switched off may be explained by considering the evolution equation of Y_s which is the source term in the above equation. Introducing steady-state values corresponding to the pump conditions, the time evolution for Y_s at short times following the pump switch off is given by

$$\dot{Y}_s = -\xi(Y_s^e - U^e X_s^e) = \tfrac{1}{2}\xi X_p X_{sp}^e\ .$$

As ξ and X_p are positive, the sign of $\dot{Y}_s$ depends only on that of the two-photon coherence in steady pump conditions Y_{sp}^e. In the above equations the superscript e denotes the equilibrium values in presence of far-ir field. As

$$Y_{sp}^e = \frac{2X_s^e}{3\eta X_p}\left[\frac{X_p^2}{2} - 1 - (X_s^e)^2\right]$$

the overshot appears if $\dot{Y}_s$ is positive, i.e., if

$$\tfrac{1}{2}X_p^2 > 1 + (X_s^2)^2\ .$$

This confirms the experimental results indicating that it appears in strong-saturation conditions.

The differential system (11) may also be solved numeri-

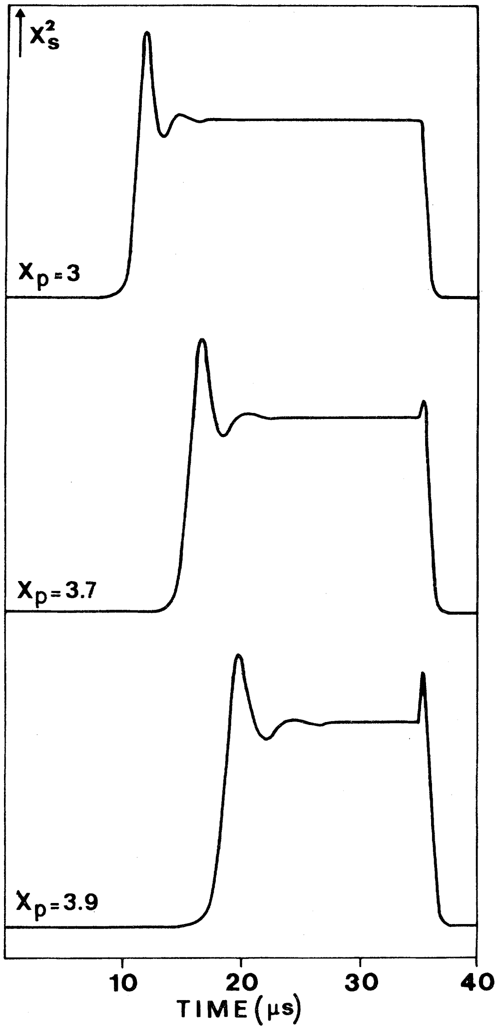

FIG. 7. Calculated transient signals using the three-level Eqs. (11) with $\eta=4$ and for various values of the pump field X_p.

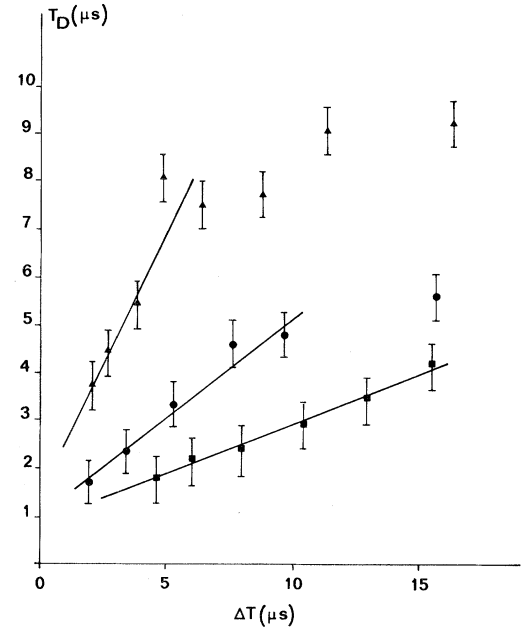

FIG. 8. Variation of the delay (T_D) between the creation of the population inversion and the onset of far-ir power as a function of the interruption time (ΔT) between the two pump pulses (▲: 22 mTorr. ●: 11 mTorr. ■: 7 mTorr).

cally. It is necessary to introduce an effective gain parameter η taking into account the Doppler broadening of the infrared transition. Figure 7 reports the results of computer calculation of the far-ir output power for different values of the ir field, always using the same average gain parameter ($\eta=4$). They show that the overshot appears in the conditions mentioned above, i.e., very strong pump saturation.

In addition to assigning the transients at the fall of laser emission, the three-level model may be used if one wants to consider the effect of Stark splitting of the pump transition or similar coherence effects on the various transients regimes observed at the onset of laser action. However, as mentioned in Sec. III, the two-level model seems adequate for describing the onset of the far-ir laser radiation in most cases.

The stability analysis used in the two-level model may be extended to Eqs. (11). However, the stability conditions obtained in that case are very heavy and their physical meaning is difficult to work out.

VI. DEPENDENCE ON THE INITIAL INTENSITY

In the semiclassical theory of the laser, the time delay T_D between the creation of the population inversion and the rise of far-ir laser power depends not only on the gain in the laser but also on the intensity I_0 present inside the laser at the beginning of the process. In usual conditions, this intensity comes from blackbody radiation and it is difficult to vary it.

In the experiments described hereafter, laser action does not start on this incoherent field but on the amount of coherent radiation which is leftover from a preceding pulse. These experiments are also of interest when the statistical properties of laser radiation buildup are considered. In fact, not only the average intensity varies but also its statistical properties since in a single pulse experiment, the number of photons of the initial field follows Bose-Einstein statistics while the residual coherent field is described by Poisson statistics.[38] The investigation of statistical properties is left for a future paper and we concentrate here on the average properties and, in particular, on the variation of the time delay T_D.

Figure 8 illustrates the dependence of T_D on the interruption time T separating the two pump pulses. The high-pressure curve (22 mTorr) shows two zones: (i) If ΔT is larger than 6 μs, T_D is independent of ΔT; the second emission is not related to the first one since the residual coherent field is weaker than blackbody radiation. (ii) If ΔT is less than 5 μs the delay varies linearly with ΔT, indicating that the far-ir laser starts on the tail of the first emission.

Experimental results clearly show that increasing initial intensity decreases the delay observed for the second emission buildup and that this delay is a linear function of the interruption time. The slope of the curves T_D vs ΔT also varies linearly with pressure with a typical value of 0.9 at 22 mTorr.

As the time delay depends on the logarithm of the initial intensity, the linear dependence of T_D vs ΔT is simply explained if the damping of the residual radiation is exponential. As shown in Sec. V, this is a rather crude approximation which is not valid at short times when the two-photon transients have to be considered. However, it may be regarded as reasonably good here since the delay experiments were carried out for long interruption times when both ir and far-ir fields are extremely weak.

VII. CONCLUSION

In this paper, we have presented the results of a general investigation of the transient regimes of a far-ir laser whose pump radiation is suddenly switched on or off. In addition to the usual "aperiodic" regime which was observed in other lasers, we have demonstrated that under strong-saturation conditions the laser output exhibits damped oscillations. These oscillations may be explained in a dynamic model of the laser in which the Bloch-Maxwell equations are solved using the "rate-equation approximation."

As the pump is stopped, the transient regimes result from the combination of the pump and the far-ir laser transients. At low pressures it was possible to observe a two-photon (Raman) transient which causes an increase of far-ir laser intensity as the pump is switched off.

The results reported here are a first step in the investigation of the far-ir laser as a synergetic system. In particular, it has been possible to observe relaxation oscillations under cw excitation conditions, which is of particular interest in regard to the recent search for the onset of chaos in lasers through period doubling transitions.[28,39] It would also be useful to go forward in the direction of more quantitative comparison between experiments and the models proposed for their interpretation as was done for lasers containing saturable absorbers.[40] The investigation of photon statistics in double pulse experiments is another development of this work. They can provide information on the influence of photon statistics as laser action sets up since they allow one to start the laser either on the blackbody radiation or on the residual coherent radiation left from the first pulse experiment.

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