Very Short Far-Infrared Pulses from Optically Pumped $\text{CH}_3\text{OH}/\text{D}$, CH_3F , and HCOOH Lasers Using an EQ -Switched CO_2 Laser as a Pump Source

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Abstract—Very short far-infrared (FIR) pulses were generated from optically pumped CH_3F , $\text{CH}_3\text{OH}/\text{D}$, and HCOOH lasers using a Q -switched, current pulsed (200 mA, 100 μs), low pressure (20 torr) CO_2 laser as a pump source. Values of 20 ns for the rise time and 50 ns for the decay time of the FIR pulses have been observed. The dependence of the FIR pulse shape parameters, i.e., rise time, decay time, and pulse buildup time, on the width of the pump pulse and the pressure of the molecular gas have been investigated experimentally. Due to the regular pulse shape, high repetition rate (350 Hz), high peak power (≥ 1 W), and broad spectral range ($\lambda = 100\text{--}500\ \mu\text{m}$), the pulses are very useful for purposes of solid-state and molecular time resolved spectroscopy.

I. INTRODUCTION

THIS paper describes the generation of far-infrared pulses, with a rise time of the order of 10^{-8} s, from optically pumped CH_3OH , CH_3OD , HCOOH , and CH_3F lasers using a Q -switched current pulsed (EQ -switched) CO_2 laser as a pump source for purposes of time resolved and saturation spectroscopy in solid-state and molecular physics.

In most reports [1] on the generation of very short optically pumped far-infrared (OPFIR) lasers, the TEA- CO_2 laser has been used as a pump source. OPFIR laser pulses as short as 10^{-9} s were generated from CH_3F and D_2O lasers by means of synchronous pumping with a mode-locked TEA- CO_2 laser [2]. Due to its broad bandwidth of several gigahertz, a TEA- CO_2 laser is an excellent pump source for off-resonance optical pumping [3] and Raman-type processes [4]. However, it has been shown that its broad bandwidth is a disadvantage for pumping other molecules, like CH_3OH and CH_3OD [5]. Pulsed emission from these molecules could only be obtained after the TEA- CO_2 laser oscillation bandwidth was narrowed down to the absorption bandwidth (~ 70 MHz) of the pump transition of these molecules [5]. In addition, the low repetition rate (~ 10 Hz) and the nonreproducible irregular pulse shape make the OPFIR pulses generated by a TEA- CO_2 laser unfavorable for sampling techniques and pulse shape analysis and therefore for time resolved spectroscopy.

Despite its high repetition rate and reproducible regular pulse

shape, the Q -switched CO_2 laser has been used only in a few cases [6], [7] as a pump source for generation of OPFIR laser pulses. The reason for this may be that for application of the Q -switching technique the gain of a conventional low pressure (~ 20 torr), the low current (~ 20 mA) CO_2 laser is only sufficient near the center of the P - and R -branches of the CO_2 spectrum.

In this paper it will be shown that due to the high current density (~ 300 mA) in the pulsed discharge of the CO_2 laser in combination with the Q -switching technique, peak powers in excess of 1 kW can easily be obtained for practically all transitions in the P - and R -branches of the CO_2 spectrum. Due to this pump source, 19 different OPFIR pulsed laser lines have been observed with peak powers in excess of 1 W and rise times of a few times 10^{-8} s. The pulses have a high repetition rate (~ 350 Hz), a regular pulse shape, a moderate peak power (≥ 1 W), and cover a broad FIR wavelength region. From the observed dependence of the FIR pulse shape parameters on the CO_2 pulse shape and the FIR gas pressure, it is concluded that values for the rise time, decay time, and width of the FIR pulses of $\leq 10^{-8}$ s are within reach of an EQ -switched CO_2 laser pumped FIR laser.

II. EXPERIMENTAL SETUP AND RESULTS

A schematic diagram of the experimental setup is shown in Fig. 1. The pump source consists of a conventional tunable low pressure (~ 20 torr) CO_2 laser which is Q -switched by a rotating mirror on an air driven spindle. Discharge current pulses up to 300 mA and of 2–200 μs duration are excited and synchronized with an adjustable delay to the Q -switch by means of a trigger pulse, a pulse delay and timer circuit, and an HV switching unit. The trigger pulse is derived from the rotating mirror, an He-Ne laser beam and an HP 4207 p-i-n diode detector. The pump pulses are focused by an $f = 50$ cm ZnSe lens onto the 1 mm coupling hole of the input mirror of the 1 m long FIR waveguide laser. Both quartz and metallic waveguides were used with diameters 13 and 25 mm. The FIR pulses transmitted through the 2 mm coupling hole of the output mirror were detected with a liquid He-cooled n-GaAs detector of which the response time is limited to 5 ns by the electronic circuit and the cable impedance. Very strong pulses could be observed with a calibrated ($125\ \mu\text{V}/\text{W}$) pyroelectric detector with a response time of 5 ns. The detector signals were amplified and recorded with a PAR 162 double boxcar integrator or a Tektronix 7834 storage oscilloscope. Via an AR-coated Ge plate,

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Fig. 1. Schematic diagram of the experimental setup for the generation of very short optically pumped far-infrared laser pulses using an *EQ*-switched CO₂ laser as a pump source.

Fig. 2. Observed peak powers for the transitions of the 9P-branch of the CO₂ spectrum for three different repetition rates (118, 215, and 325 Hz), and for both the *Q*-switched and *EQ*-switched mode of operation.

part of the CO₂ pulse (~ 2 percent) is monitored by a liquid N₂-cooled PbSnTe detector with a rise time of 5 ns.

The peak powers observed for the transition of the 9P-branch of the CO₂ spectrum are shown in Fig. 2 for three different repetition rates and for both the *Q*-switched (open bars, ~ 15 mA dc) and *EQ*-switched (solid bars, 150 mA, 100 μ s) mode of operation. Similar diagrams were obtained for the 9R, 10P, and 10R branches. It is evident that operation in the *EQ*-switched mode improves both the peak power and the repetition rate of the pulses substantially with respect to the *Q*-

Fig. 3. The observed *EQ*-switched pulse shape of the 9P20 transition at a repetition rate of 324 Hz and for three different values of the delay t between the start of the discharge current pulse and the laser pulse. The inset shows a discharge current pulse of 200 mA and a duration of 40 μ s followed by the corresponding laser pulse.

switched mode of operation. Peak powers in excess of 1 kW and repetition rates up to 325 Hz can be obtained even at transitions such as 9P38, 9P36, 9P6, and 9R6, which are especially important for pumping CH₃OH and CH₃OD. It should be noted that the results of Fig. 2 give only a defective view of the maximum attainable values for the peak power since the 25 percent transmission of the out-coupling mirror implies that the cavity is undercoupled for the stronger lines. By a suitable choice of the repetition rate and the transmission of the outcoupling mirror, pulse powers in excess of 10 kW may easily be generated for all lines in the 9P-branch. It should further be noted that for the very strong lines still higher repetition rate may increase the peak power (and decrease the pulsewidth). However, the repetition rate of our system is limited to 350 Hz by mechanical instabilities of the rotating mirror.

In the case of a conventional *Q*-switched CO₂ laser, the pulsewidth is determined by the gain of the transition and the repetition rate of the rotating mirror. The higher the gain, the higher the repetition rate for maximum power, and the shorter the pulsewidth. In the case of an *EQ*-switched CO₂ laser, the pulsewidth is, in addition, determined by the delay t between the start of the discharge current pulse and the laser pulse. Fig. 3 shows the observed pulse shape of the 9P20 transition at a repetition rate of 324 Hz and for different values of delay t . The inset shows the discharge current pulse followed by the corresponding laser pulse. The highest output power was obtained for $t = 72 \mu$ s and for a current pulse of 200 mA and 40 μ s duration. The long delay is due to the buildup time of the population inversion in the CO₂ laser levels involved. Increasing the current pulse duration has no effect on the laser pulse because of saturation at the operating CO₂ pressure (~ 3 torr). Due to the long buildup time of $\sim 100 \mu$ s, the maximum repetition rate is limited to ~ 10 kHz. Finally, it follows from Fig. 3 that by variation of t the peak power and the width of the laser pulse can be affected.

Table I gives a survey of the 19 observed *EQ*-switched pumped laser lines in CH₃OH, CH₃OD, HCOOH, and CH₃F and some

TABLE I
SURVEY OF THE 19 OBSERVED EQ-SWITCHED CO₂ LASER PUMPED LASER
LINES IN CH₃OH/D, HCOOH, AND CH₃F AND SOME OF THEIR
CHARACTERISTICS

Molecule	CO ₂ pumpline	Wavelength (μ m)	Optimum pressure (mTorr)	Peak power (W)	Pulsewidth (ns)	Risetime (ns)	Decaytime (ns)
CH ₃ OH	9P34	70.6	480	5	200	80	270
		204 ^o	520	5	60	45	60
	9P36	118.8	800	>10	100	40	120
	9P38	193.2	480	5	170	40	170
		198.8	480	5	170	40	170
	10R16	62 ^x	250	<1	-	-	-
		78	250	<1	-	-	-
		208 ^x	250	<1	-	-	-
CH ₃ OD	9P6	134.7	250	5	100	40	100
		229.1	250	5	80	40	110
	9P10 ^{xx}	133 ^x	450	5	90	50	90
	9P30	103	825	>10	120	45	150
	9R6 ^{xx}	282 ^x	270	>10	55	20	60
		57	825	5	128	48	128
CH ₃ F	9P20	496	1500	>10	70	25	60
HCOOH	9R28	516	400	5	200	80	200
	9R24	420	450	5	160	52	250
	9P18	396	400	5	150	48	240
	9R20	426	350	5	120	36	240

x New FIR line

xx New pumpline

o Observed with dc Stark effect

of their characteristic parameters. Four of these lines (indicated by an asterisk) have not been reported before, while one line at $\lambda = 204 \mu\text{m}$ in CH₃OH was observed under application of an electric field to the FIR cavity [8]. The wavelength of the pulsed lines were obtained by scanning the FIR cavity length while monitoring the chopped output with a Golay cell. The peak powers of the strongest FIR pulses (≥ 10 W) were determined from measurements with a calibrated pyroelectric detector, while those for the weaker lines were obtained from relative measurements with a Golay cell. The pulse shape parameters, i.e., rise time, width, and decay time, were determined from the pulse shape measurements with the He-cooled n-GaAs detector. The values vary with the pump line due to different peak power duration and energy of the pump pulse; with the pump and laser transition in the molecular gas due to the different radiation- and collisional-relaxation rates. The observed values are typically 40 ns for the rise time and 150 ns for both the width and the decay time: for $\lambda = 204 \mu\text{m}$ in CH₃OH, $\lambda = 282 \mu\text{m}$ in CH₃OD, and $\lambda = 496 \mu\text{m}$ in CH₃F, the rise time is as short as 20 ns, the pulsewidth and the decay time 50 ns. From the data in the table and those presented in Fig. 2, an energy conversion efficiency (ratio of the FIR pulse energy to the CO₂ pulse energy) of the order of 10^{-3} was obtained for the strongest laser lines. This number corresponds to approximately 10 percent of the theoretical quantum efficiency and is comparable with the conversion efficiency of the best CW systems.

Fig. 4 shows a storage oscilloscope picture of a single very short FIR pulse at $\lambda = 282 \mu\text{m}$ in CH₃OD with a rise time of

Fig. 4. Storage oscilloscope picture of a single FIR pulse at $\lambda = 282 \mu\text{m}$ in CH₃OD with a rise time of 20 ns and a decay time of 50 ns.

$2 \cdot 10^{-8}$ s and for both the pulsewidth and decay time a value of $5 \cdot 10^{-8}$ s. Due to the regular pulse shape and the very good signal-to-noise ratio, these pulses may be very useful for purposes of solid-state and molecular time resolved spectroscopy.

III. DISCUSSION

The FIR pulses are characterized by the rise time t_r , the decay time t_d , and the pulsewidth Δt ; furthermore, there is a pulse buildup time t_b , which is defined as the time between the start of the pump pulse and the observable onset (~ 5 percent of its peak power) of the FIR laser pulse. t_r and t_d are defined as the time between 10 and 90 percent of the peak power at, respectively, the leading and trailing edge of the pulse Δt as the full width at half maximum. Both t_r and t_b depend on the gain $g(\nu) = Bh\nu\Delta N_{\text{FIR}}$, where B is the FIR transition rate, $h\nu$ is the FIR quantum, and ΔN_{FIR} is the population inversion density which is built up during the time t_b , on

Fig. 5. The observed dependence of the FIR pulsewidth on the width of the CO₂ laser pulse for laser lines at $\lambda = 103$, 133, and 282 μm in CH₃OD.

the intensity and width of the pump pulse, and on the pressure of the molecular gas. After the onset of the rapidly increasing FIR pulse, ΔN_{FIR} is quickly depleted and the gain is decreased with respect to the initial small-signal gain.

The decay time depends on the molecular rotational relaxation time $(\pi\Delta\nu)^{-1}$ where $\Delta\nu$ is the homogeneous linewidth of the laser transition, which is approximately 40 MHz at 1 torr for the polar laser molecules that have been studied to date [9]. If the FIR pulse falls within the duration of the pump pulse, the tail of the pump pulse will continue to pump the FIR gas and have an effect on t_d . The width of the FIR pulse depends on both the rise time and decay time and therefore on the characteristics of the pump pulse and the pressure of the molecular gas.

Fig. 5 shows the observed dependence of the FIR pulsewidth on the width of the CO₂ laser pulse for several laser lines at $\lambda = 103$, 133, and 282 μm in CH₃OD. The CO₂ laser pulsewidth was varied by means of the repetition rate of the rotating mirror; the amplitude of the peak was held constant at a value of a few kilowatts. The linear relation between the two pulsewidths is typical for most of the observed FIR pulses and shows that one may expect a smaller FIR pulsewidth for a smaller CO₂ pulsewidth.

Fig. 6 presents values of the different pulse shape parameters as a function of pressure of the molecular gas for $\lambda = 496 \mu\text{m}$ in CH₃F and $\lambda = 282 \mu\text{m}$ in CH₃OD. The results were obtained from observations of the FIR pulse shapes and the corresponding pump pulses as a function of pressure, which are shown in Fig. 7. The results for $\lambda = 496 \mu\text{m}$ in CH₃F show that both t_b and t_r are constant over the total pressure range, which indicates that the gain $g(\nu)$ must be a constant of the pressure as was shown previously [7]. The decay time t_d changes from ~ 130 ns at 350 mtorr to ~ 90 ns at 1200 mtorr. Since Fig. 7 shows that for all pressures the FIR pulses at $\lambda = 496 \mu\text{m}$ fall completely within the width of the CO₂ pulse, the change in t_d can be only due to the variation of the rotational relaxation time $(\pi\Delta\nu)^{-1}$ with pressure. For $\lambda = 282 \mu\text{m}$ in CH₃OD pulse buildup time, rise time and decay time are practically constant

Fig. 6. Observed values for the rise time (t_r), decay time (t_d), and the pulse buildup time (t_b) as a function of the pressure of the molecular gas for $\lambda = 496 \mu\text{m}$ in CH₃F and $\lambda = 282 \mu\text{m}$ in CH₃OD.

Fig. 7. Observed FIR pulse shapes as a function of pressure of the molecular gas and the corresponding pump pulse for $\lambda = 282 \mu\text{m}$ in CH₃OD and $\lambda = 496 \mu\text{m}$ in CH₃F.

from 450–1400 mtorr indicating that also for this line the gain is constant and the decay time is mainly determined by the width of the pump pulse. For $p < 450$ mtorr, t_b increases so that for $p = 350$ mtorr, the FIR pulse is completely after the pump pulse. The experimental value of $t_d = 50$ ns at $p = 350$ mtorr is in rather good agreement with the calculated value of the molecular rotational relaxation time $(\pi\Delta\nu)^{-1}$, which is ~ 30 ns at $p = 350$ mtorr. It should further be noted that t_r is constant down to 350 mtorr while t_b increases remarkably. This may be due to the fact that t_b depends on the small-signal gain which is proportional to the pressure of the molecular gas, while t_r is determined by the gain in presence of an intensive radiation field.

So, in conclusion, it follows from the results of Figs. 5-7 that variation of the width of the pump pulse and the pressure of the molecular gas has the most dramatic effect on the decay time and therefore on the width of the FIR pulse. Due to the limitations of our experimental setup, the present CO₂ pulse tail is limited to at least 100 ns, so that for pressures of $p \geq 0.1$ torr, the decay time and the width of the FIR pulse are mainly determined by the width of the pump pulse. However, *Q*-switched CO₂ laser pulses with a width of 20 ns have been generated using an intracavity CdTe electrooptic modulator [10]. It is reasonable to expect that optical pumping with these pulses will give rise to FIR pulses of which the ultimate linewidth in the high pressure limit is determined by the cavity lifetime of the FIR radiation [11], which is of the order of 20 ns for our system.

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