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Very Short Far-Infrared Pulses from Optically Pumped CH$_3$OH/D, CH$_3$F, and HCOOH Lasers Using an $Q$-Switched CO$_2$ Laser as a Pump Source

H. J. A. BLUYSSSEN, A. F. VAN ETTEGER, J. C. MAAN, AND P. WYDER

Abstract—Very short far-infrared (FIR) pulses were generated from optically pumped CH$_3$OH, CH$_3$OD, and HCOOH lasers using a $Q$-switched, current pulsed (200 mA, 100 $\mu$s), 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 ($\geq$1 W), and broad spectral range ($\lambda$ = 100-500 $\mu$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$_3$OH, CH$_3$OD, HCOOH, and CH$_3$F 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$_3$F and D$_2$O 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$_3$OH and CH$_3$OD [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 ($\sim$70 MHz) of the pump transition of these molecules [5]. In addition, the low repetition rate ($\sim$10 Hz) and the nonreproducible irregular pulse shape make the OPFIR pulses generated by a TEA-CO$_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 ($\sim$20 torr), the low current ($\sim$20 mA) CO$_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 ($\sim$300 mA) in the pulsed discharge of the CO$_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 ($\sim$350 Hz), a regular pulse shape, a moderate peak power ($\geq$1 W), and cover a broad FIR wavelength region. From the observed dependence of the FIR pulse shape parameters on the CO$_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 ($\sim$20 torr) CO$_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 $\mu$s 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 $\mu$W/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,
Recorder

Copulseshapes

Fig. 1. Schematic diagram of the experimental setup for the generation of very short optically pumped far-infrared laser pulses using an EQ-switched CO$_2$ laser as a pump source.

Fig. 2. Observed peak powers for the transitions of the 9P-branch of the CO$_2$ spectrum for three different repetition rates (118, 225, and 325 Hz), and for both the Q-switched and EQ-switched mode of operation.

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 $\mu$s followed by the corresponding laser pulse.

The peak powers observed for the transition of the 9P-branch of the CO$_2$ spectrum for three different repetition rates (118, 225, and 325 Hz), and for both the Q-switched and EQ-switched mode of operation.

Table I gives a survey of the 19 observed EQ-switched pumped laser lines in CH$_3$OH, CH$_3$OD, HCOOH, and CH$_3$F and some
TABLE I

<table>
<thead>
<tr>
<th>Molecule</th>
<th>CO₂ pumpline</th>
<th>Wavelength (μm)</th>
<th>Optimum pressure (mTorr)</th>
<th>Peak power (W)</th>
<th>Pulsewidth (ns)</th>
<th>Risetime (ns)</th>
<th>Decaytime (ns)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CH₃OH</td>
<td>9P₃</td>
<td>72.6</td>
<td>480</td>
<td>5</td>
<td>200</td>
<td>80</td>
<td>270</td>
</tr>
<tr>
<td></td>
<td>204°C</td>
<td>520</td>
<td>5</td>
<td>60</td>
<td>45</td>
<td>50</td>
<td></td>
</tr>
<tr>
<td>CH₃OD</td>
<td>9P₉</td>
<td>118.8</td>
<td>800</td>
<td>&gt;10</td>
<td>100</td>
<td>40</td>
<td>120</td>
</tr>
<tr>
<td></td>
<td></td>
<td>193.2</td>
<td>480</td>
<td>5</td>
<td>170</td>
<td>40</td>
<td>170</td>
</tr>
<tr>
<td></td>
<td>198.8</td>
<td>480</td>
<td>5</td>
<td>170</td>
<td>40</td>
<td>170</td>
<td></td>
</tr>
<tr>
<td></td>
<td>1106°F</td>
<td>62°C</td>
<td>256</td>
<td>&lt;1</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td></td>
<td>78</td>
<td>256</td>
<td>&lt;1</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td></td>
<td>206°C</td>
<td>256</td>
<td>&lt;1</td>
<td>-</td>
<td>-</td>
<td>-</td>
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<tr>
<td>HCOOH</td>
<td>9P₂</td>
<td>133.7</td>
<td>650</td>
<td>5</td>
<td>100</td>
<td>40</td>
<td>100</td>
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<tr>
<td></td>
<td></td>
<td>125.1</td>
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<td>80</td>
<td>40</td>
<td>110</td>
</tr>
<tr>
<td></td>
<td>9P₂₉</td>
<td>133°C</td>
<td>650</td>
<td>5</td>
<td>90</td>
<td>40</td>
<td>90</td>
</tr>
<tr>
<td></td>
<td></td>
<td>103</td>
<td>825</td>
<td>&gt;10</td>
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<td>40</td>
<td>150</td>
</tr>
<tr>
<td></td>
<td>262°C</td>
<td>270</td>
<td>&gt;10</td>
<td>120</td>
<td>40</td>
<td>40</td>
<td></td>
</tr>
<tr>
<td></td>
<td>57</td>
<td>925</td>
<td>5</td>
<td>120</td>
<td>40</td>
<td>40</td>
<td></td>
</tr>
<tr>
<td>CH₃F</td>
<td>9P₂₀</td>
<td>496</td>
<td>1500</td>
<td>&gt;10</td>
<td>70</td>
<td>25</td>
<td>60</td>
</tr>
<tr>
<td></td>
<td></td>
<td>516</td>
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<td>5</td>
<td>100</td>
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<td>200</td>
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<tr>
<td></td>
<td>420</td>
<td>400</td>
<td>5</td>
<td>160</td>
<td>52</td>
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<td></td>
</tr>
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<td></td>
<td>396</td>
<td>400</td>
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<td>150</td>
<td>48</td>
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<td></td>
</tr>
<tr>
<td></td>
<td>426</td>
<td>350</td>
<td>5</td>
<td>120</td>
<td>36</td>
<td>240</td>
<td></td>
</tr>
</tbody>
</table>

**Notes:**
- * New FIR line
- **XX** New pumpline
- ° 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 \( \lambda = 204 \mu m \) 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 (\( \geq 10 \) 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 \( \lambda = 204 \mu m \) in CH₃OH, \( \lambda = 282 \mu m \) in CH₃OD, and \( \lambda = 496 \mu m \) 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. Storage oscilloscope picture of a single FIR pulse at \( \lambda = 282 \mu m \) in CH₃OD with a rise time of 20 ns and a decay time of 50 ns.](image)

2 \( \cdot 10^{-8} \) s and for both the pulsedwidth 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 pulsedwidth \( \Delta t \); 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 \( \Delta t \) as the full width at half maximum. Both \( t_r \) and \( t_b \) depend on the gain \( g(\nu) = B h \nu \Delta N_{\text{FIR}} \), where \( B \) is the FIR transition rate, \( h \nu \) is the FIR quantum, and \( \Delta N_{\text{FIR}} \) is the population inversion density which is built up during the time \( t_b \), on.
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, $\Delta N_{\text{FIR}}$ 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 CO$_2$ laser pulse for several laser lines at $\lambda = 103, 133, \text{ and } 282 \mu m$ in CH$_3$OD. The CO$_2$ 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 CO$_2$ pulsewidth.

Fig. 6 presents values of the different pulse shape parameters as a function of pressure of the molecular gas for $\lambda = 496 \mu m$ in CH$_3$F and $\lambda = 282 \mu m$ in CH$_3$OD. 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 $\lambda = 496 \mu m$ in CH$_3$F 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 $\sim 130 \text{ ns}$ at 350 mtorr to $\sim 90 \text{ ns}$ at 1200 mtorr. Since Fig. 7 shows that for all pressures the FIR pulses at $\lambda = 496 \mu m$ fall completely within the width of the CO$_2$ 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 $\lambda = 282 \mu m$ in CH$_3$OD pulse buildup time, rise time and decay time are practically constant 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 $p < 450 \text{ mtorr}$, $t_b$ increases so that for $p = 350 \text{ mtorr}$, the FIR pulse is completely after the pump pulse. The experimental value of $t_d = 50 \text{ ns}$ at $p = 350 \text{ mtorr}$ is in rather good agreement with the calculated value of the molecular rotational relaxation time $(\pi \Delta \nu)^{-1}$, which is $\sim 30 \text{ ns}$ at $p = 350 \text{ mtorr}$. 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 $t_r$ 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 CO$_2$ 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 CO$_2$ 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 tail is limited to at least 100 ns, so that for pressures of 20 ns for our system.

**REFERENCES**


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