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Author(s):

G.E. Busch, C. J. Hewitt, D. K. Remelius,
T. Shimada and D. C. Thompson

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G.E. Busch, C. J. Hewitt, D. K. Remelius, T. Shimada and D. C. Thompson
Los Alamos National Laboratory, Los Alamos, NM 87545

Abstract:

We have developed rapidly tuned RF-pumped CO₂ waveguide laser transmitters for remote sensing in the 9-11 μm spectral range. The small size, high power and efficiency, and tunability of these lasers offer significant advantages over other laser sources in this spectral region. Employing acousto-optic modulators to achieve random-access tuning at pulse rates up to 100 kHz permits rapid gathering of data on time scales short compared to times for change in atmospheric turbulence and absorption effects, thereby improving the signal-to-noise ratios that can be achieved. Laser system design and performance characteristics of present systems are described, along with proposed concepts to increase optical bandwidths and extend the tuning range to cover the full long-wave atmospheric transmission window from 8-12 μm .

I. Laser Transmitter Development Background

Tunable long-wave infrared lasers are utilized for differential absorption lidar (DIAL) remote sensing in the long-wave infrared (LWIR) “fingerprint” region of the vibrational spectra of many chemicals near 10 μm . Although techniques to generate tunable radiation in this spectral region using nonlinear techniques, starting from 1 μm solid state lasers, have been under development for 30 years¹, the inefficiencies and low powers available to date render these approaches less than satisfactory for long range remote sensing. In the absence of solid-state lasers to fill this need, we have continued to develop and utilize CO₂ lasers as laser transmitters for remote sensing.^{2,3,4} While solid-state lasers are versatile and convenient for many low to medium power applications in the near and mid-infrared spectral regions, CO₂ lasers continue to be the most powerful and most efficient lasers readily available for LWIR applications. Since CO₂ lasers are also tunable over many rotational lines throughout the 9-11 μm spectral region, where many molecules have characteristic absorption, they have long been the lasers of choice for remote sensing systems⁵. The high average power capability translates into high pulse repetition rates needed to reduce the noise from time-dependent atmospheric turbulence, absorption, and laser speckle. In what follows, the characteristics of high-speed tunable CO₂ lasers under development are described. Ongoing work on the enabling technology of acousto-optic (AO) modulators and deflectors for fast laser tuning and system diagnostics is then presented, followed by recent work resulting in substantially improved waveguide performance. Finally, suggestions are made for future approaches to extend the tuning range and increase the bandwidth covered by LWIR lasers by combining CO₂ laser pump technology with nonlinear conversion techniques.

II. 100 kHz Random-Access-Tuned Q-switched CO₂ Lasers

Radio Frequency (RF) pumped waveguide CO₂ lasers have been shown to be compact, robust, reliable devices for industrial and laboratory applications⁶. Efficiencies approaching 10 % are generally achievable, and maintenance-free sealed-off operation with gas lifetimes approaching 20,000 hours have become routine. The beam quality provided by the waveguide mode is usually excellent, with typical $M^2 \sim 1.2$. The low voltages permitted by a transverse pumping geometry reduce the complications often encountered with the high voltages required for longitudinal discharge gas lasers. Remote sensing systems employing RF-pumped CO₂ have been operated at high pulse repetition and tuning rates in the range of 5-10 kHz with high reliability and good performance⁷. In the present paper, we describe development efforts to extend the pulse and tuning rates to ultra-high ranges near 100 kHz,⁴ and describe technologies that are being developed to enhance the performance and wavelength ranges covered by LWIR remote sensing laser systems. Figure 1 shows a block diagram of the computer controlled 100 kHz randomly tuned Q-switched CO₂ laser system. The modified DEOS NV laser head gain cell shown in Fig. 1 consists of a 2.8 mm square cross section BeO waveguide, which has a total discharge gain length of 220 cm, folded into 5 sections, with fold mirrors at each vertex. The waveguide gain cell is pumped with continuous RF power, but laser

power is extracted in pulses. Individual rotational lines of CO₂ are selected for oscillation, under computer control, by changing the RF frequency to the AO modulators (AOMs) in the cavity. By pulsing and controlling the temporal width of the RF signal to the AOMs, the laser produces a Q-switched short pulse, with pulse length determined by both the laser gain and the falling edge of the RF pulse. An intra-cavity cylindrical telescope expands the mode size in the AOMs to achieve the resolution for single line operation.

Fig. 1. Schematic layout of A/O tuned CO₂ laser

Typical temporal behavior of the laser pulses when the laser system is operated in an 11-line sequence (10P(J=10-30)) at 100 kHz pulse rate is presented in Fig. 2. The small variations in the oscilloscope waveform, from sequence to sequence, are artifacts caused by sampling rates that are not synchronous with laser pulse repetition rates, rather than being real power variations. Average power was measured to be 8 W.

Fig. 2. Oscilloscope trace of 11-line sequence (10P(J=10-30)) at a pulse rate of 100 kHz

Long-term laser stability was investigated further under conditions similar to those in Fig. 3, except that the data acquisition system speed limited the pulse repetition rate to 50 kHz, instead of 100 kHz. Data displayed in Fig. 3 for the 10P(20) line was taken with a mercury cadmium telluride (MCT) detector and a boxcar integrator synchronized with the 10P(20) line trigger source from the controller. Laser stability over a 10-minute measurement interval was excellent, having an rms deviation of 1.36%. Similar data for the 10P(30) line had stability degraded to 2.76%, due mainly to lower small signal gain. No dropouts were observed at either line during the 10-minute sampling period. It is expected that the stability on all lines will be improved with better temperature control of the laser coolant.

Fig. 3. Pulse to pulse laser stability at 10P(20) during 11-line sequence operation with a pulse repetition rate of 50kHz.

III. Acousto-Optic Laser Tuning

Many of the theoretical and practical details of AO tuning of CO₂ lasers have been previously published.^{2,3} The simple configuration shown in Fig. 1 uses a pair of AOMs, each operating at a fixed Bragg angle θ_B . This gives highly efficient diffraction of the incident laser beam into a single diffraction order when the (RF) acoustic frequency ν_a and free-space optical wavelength λ_0 satisfy the Bragg relation:

$$\sin \theta_B = \frac{\lambda_0 \nu_a}{2nV_a}$$

in which n and V_a are the index of refraction and acoustic velocity in the AO crystal. The acoustic frequency then determines the selected wavelength. All other wavelengths are diffracted at the wrong angle to resonate in the laser cavity. Two modulators, with oppositely directed acoustic waves, are required to cancel the optical frequency shifts arising from diffraction from moving acoustic waves. Without this cancellation, the net frequency shift in sequential round trips would be additive, and for narrow laser gain bandwidths, as is the case for CO₂, could prevent oscillation entirely after only a few round trips. The afocal telescope improves resolution by expanding the laser mode entering the modulator, to illuminate a greater number of lines of the acoustic grating.

We are working with industry to develop improved tuner technology. The developmental devices have operated at frequencies up to 200 MHz, with transducer heights as small as 0.5 mm. Brewster angle entrance and exit faces provide anamorphic beam expansion. We have made interferometric measurements at wavelengths near 10 μ m, such as that shown in Fig. 4, which provide important information about thermal gradients in these devices, and we have developed models to further elucidate important parameters

Fig. 4. Panel a: Measured interferogram at 10 μ m for the current AO tuner at high repetition rate. Dominant heat sources are the acoustic transducer and absorber at the ends of the Ge AO crystal, while heat removal is accomplished by edge cooling at the top and bottom. A temperature difference of 1°C gives rise to a single-pass optical phase

difference of ~ 1.2 waves in each AO crystal. Panel b: Constant temperature surfaces within the AO tuner, from analysis of the interferogram in panel a.

and to help guide designs of future devices. The models include many physical effects, including angular and spatial overlap between optical and acoustic beams; angular mismatch due to the frequency shift; the effect of finite interaction length; linear and non-linear acoustic attenuation in the crystal; acoustic diffraction; optical absorption; and thermal effects from bulk absorption of acoustic and optical power. Many useful conclusions are derived from the models, among them being that higher AO frequencies and smaller transducer heights are advantageous. Fig. 5 shows contour plots of modeled tuner efficiency versus acoustic frequency and interaction length. Both our measurements and our model have clearly demonstrated the importance of careful thermal management for high duty factor AO devices to minimize optical distortion arising from thermal gradients.

Fig. 5. Contour plots of overall tuner efficiency versus acoustic frequency and interaction length for (a) 6-mm transducer height and (b) minimum transducer height. The plots are for a Germanium device at $10\ \mu\text{m}$, with the optical beam width as required for a fixed spectral resolution of $0.75\ \text{cm}^{-1}$. The solid dot in (a) predicts the performance with particular commercial devices (Isomet 1208-6).

Our long-term goal is a compact integrated tuner, with both of the AO modulators and the high reflector integrated into a single monolithic device. Such a device would readily be incorporated in laser heads, providing a rugged, flexible and high-performance tuner for field applications.

IV. Fast Acousto-Optic Diagnostics

Figure 6 shows the system diagnostics for characterizing and maintaining the fielded LANL lidar system. A pair of uncoated ZnSe wedges at near Brewster's angle split off a few percent of the light for diagnosing the transmitter performance. The reflection from the first wedge enters a far-field spatial diagnostic. The camera (Spiricon Pyrocam II) operates at frame rates of up to 100 Hz, which is far too slow to capture single pulses, as the laser is being rapidly tuned through 40 or more lines, at the repetition rate of the laser. Since

Fig. 6. Schematic diagram of diagnostics for field lidar system. Two near-Brewster-angle wedges send small fractions of the transmitter output to the various diagnostics, including the pulse-picked far-field spatial diagnostic, the DIAL reference detector, a power meter and the acousto-optic spectrometer. A removable mirror can direct the entire remaining transmitter output to a power meter for cross-calibration.

measurements are needed on each laser line, by this and other slow diagnostics, a “pulse picker” enables individual pulses to be selected at any rate reduction factor required. This is accomplished by an AOM (Isomet 1208-6) operating on the same acoustic frequency as the AO laser tuner so that, as with the laser tuner, the deflection angle is constant, independent of the chosen wavelength. Use of the diffracted, rather than an undiffracted beam for diagnostics, ensures an extremely high contrast ratio for pulse selection, something that is very important when only one pulse in 40 or 50 is being selected. Also, this method can support repetition rates of hundreds of kilohertz. AO pulse picking is superior to electro-optic techniques in several respects. In addition to higher available contrast ratios and repetition rates, the use of lower voltages in AO devices results in less EMI and easier packaging for field systems.

The second wedge shown in Fig. 6 splits off light to the lidar reference detector suite, composed of a thermoelectrically cooled single-element MCT photoconductor, a power meter, and the AO spectrometer. High-speed differential absorption lidar (DIAL) requires a rapid spectral diagnostic for transmitted light, since proper analysis of the return signals depends on knowing the spectral purity of the transmitted beam. Our spectrometer, shown schematically in Fig. 7, has been previously described in some detail⁸. The device is essentially a scanned monochromator, with the usual rotating grating replaced by the combination of an AO deflector and a 75 lines/mm grating. Tuning is accomplished by varying the frequency of the RF signal fed to the piezoelectric transducers of the deflector. The deflector (Isomet LS-600) has a large 75 mm (horizontal) aperture and a 40 MHz (50-90 MHz) bandwidth, giving a tuning range of 9.0 to 11.4 μm ,

Fig. 7. Schematic of AO spectrometer, with top (above) and side views. Light focused onto the alignment pinhole expands in the horizontal direction to the collimating mirror. Light is collimated in the vertical direction by the cylindrical lens. After passing through the AO deflector (twice) and being diffracted by the grating, which also introduces a small vertical angle to separate the input and output beams, the light is focused onto a small detector by the collimating mirror and a second cylindrical lens.

and a resolution $\sim 0.005 \mu\text{m}$. Cylindrical optics enable the light to be expanded to fill the full 75 mm horizontal aperture with a narrow vertical focus to pass through the 4-mm vertical aperture of the deflector. The large aperture and the acoustic velocity limit the tuning speed to 73 kHz. Increased resolution of the spectrometer, or increased tuning speed at the same resolution, could be accomplished by altering the setup to make multiple optical passes, but throughput would also decrease, due to increased losses.

V. Improvements to Laser Waveguides

The present CO_2 laser system performance could be further enhanced for remote sensing applications with several component improvements. For example, a higher laser gain would improve the pulse energy and stability on all lines and would also increase the total number of laser lines having useful energy and stability. Alternatively, a higher gain coefficient would permit the size of the transmitter to be reduced for the same performance, increasing the variety of platforms and applications for active remote sensing.

To investigate methods that might improve gain, we have developed a flexible waveguide test-bed with adjustable waveguide dimensions, materials, and impedance matching circuits. This versatile test-bed, has enabled a large number of parameters to be varied to optimize the performance of RF-pumped CO_2 waveguide lasers. Some results of the parametric studies are presented below, followed by examples of the use of those results in predictive laser performance models to help design systems for future remote sensing platforms. These models point the way to systems with substantially better performance than obtained from presently deployed systems, in terms of laser power, pulsewidth, number of useful transition lines, output stability, and total wall plug efficiency.

A. CO_2 waveguide test-bed description and results

We have utilized the RF-pumped waveguide test-bed to investigate several experimental parameters, including

- 1) Waveguide interior transverse dimensions.
- 2) Gas mixture and pressure-dependence of performance.
- 3) Catalysis effects for possible reconstitution of discharge decomposition products.
- 4) Waveguide optical coupling efficiencies.
- 5) Waveguide transport effects from surface material and finish.
- 6) RF circuit impedance matching.

Though item 5), was not investigated in the same detail as the remaining five items, conclusions were still reached on the contributions of these parameters to waveguide losses, based on models and on investigations reported in the literature. Predictions and results from the first four investigations are summarized next.

From the earliest days of high power CO₂ lasers, it has been noted that the maximum cw average laser power per unit length of the gain medium, appears to be a constant, independent of the diameter of symmetric discharge channels. This result is consistent with a standard diffusion-cooled thermal mechanism⁹ for a uniform volumetrically heated, edge-cooled symmetric rod, in which the temperature rise at the center of the rod is independent of rod diameter for fixed power input per unit length. If performance were to always sharply begin to degrade at the same temperature, then maximum power input and output will also be independent of diameter. However, the situation changes for asymmetric rods, since transport in the direction of the smaller dimension becomes rate determining. This leads to the conclusion that a *decrease*¹⁰ in waveguide channel size in one dimension might actually *increase* the RF discharge power that can be usefully applied to a waveguide. The predictions are shown in Fig. 8, where at fixed RF power density, decreasing the channel width by a factor of 10 decreases the temperature rise by the square of the width (ignoring vertical transport), or by a factor of 100. If RF power is increased by the same factor of 100, to a level that is higher than the original total power by a factor of 10, the modeled temperature rise during excitation once again has the same value that it reaches in the symmetric waveguide.

Fig. 8. Depiction of the predicted improvement in performance possible as the cross-section of the laser waveguide changes from symmetric to asymmetric. Note the asymmetric waveguide may produce up to ten times the laser power as the symmetric case at the same center-to-edge temperature difference ΔT_0 . Modeling suggests $\Delta T_0 \sim 300$ K.

If the foregoing model were valid, then this increased pump power could increase the gain per unit length and the extracted laser power of the waveguide substantially.

Results of experiments carried out in the waveguide test-bed apparatus are displayed in Fig. 9, for several waveguide dimensions and gas densities, starting with a symmetric waveguide with a 2.25 mm square cross-section. Whereas the small-signal gain coefficient did in fact increase throughout the pressure range investigated, as the channel width was decreased from 2.25 mm to 1 mm, the factor was substantially smaller than the value of 2.25 that would have been expected for a diffusion-limited thermal transport process. Gain coefficients were substantially equal at 1 mm and 0.75 mm, but fell precipitously at 0.5mm.

B. Catalysis. A possible conclusion from the results in Fig.9 is that the thermal transport-limited model is incorrect. However, it has been observed in CO₂ lasers that a large fraction of the CO₂ becomes dissociated into CO and O₂ by two mechanisms: 1) direct electron impact dissociation and 2) dissociative electron attachment reactions in the medium. The dissociated fraction can exceed 70% under strong pumping conditions.¹¹

Fig.9. Small-signal gain coefficient vs waveguide channel width and gain medium gas pressure. Small increase is observed in the decrease in width from 2.25 to 1 mm. However this increase is almost eliminated (at least at lower pressures) with further decrease in width to 0.75 mm, and falls significantly for 0.5 mm width. This result may be partially explained by electron impact dissociation of CO_2 at high current density.

Since discharge currents were increased roughly inversely to the channel width in the experiments presented in Fig. 9, it is possible that these deleterious dissociation processes could be countering the expected improvements from increased thermal transport rates. This possibility was reinforced by recent reports¹² of substantial improvement in the performance of RF-pumped slab CO_2 lasers, using heated gold-plated discharge electrodes to catalytically recombine the CO and O_2 . As a test of the dissociation/recombination mechanisms, we replaced the bare aluminum electrode of the waveguide test-bed apparatus with a gold-plated copper electrode, and observed the marked increase in small-signal gain displayed in Fig.10. The 2.25 mm width aluminum electrode result in Fig. 9 is included for better comparison with Fig.10. While the improvement was small for the symmetric waveguide, substantial improvements are demonstrated for 0.75 and 1.00 mm channels. The pressure range over which performance of the 0.75 mm waveguide is superior to that of the 1.00 mm waveguide was also increased.

Fig. 10. Measured small signal gain dependence on waveguide channel width and gas pressure for two different electrode materials (gold and aluminum).

Further benefits are expected to accrue from the smaller waveguide geometry and higher operating pressures. First of all, the laser medium should recover faster between pulses, under these conditions, since most of the relevant kinetic processes, being second order reactions, are faster under these conditions. This is highly beneficial for high repetition-rate operation. In addition, the increased pressure increases collisional broadening of the laser transition, supplying the bandwidth to enable the laser to operate with shorter pulsewidths. This could be particularly important for mode-locked operation. Finally, for a given gain, an increased linewidth results in increased energy storage, and thus greater extractable energy.

C. Modeled performance for next generation system configuration

The model we have developed for transmitter performance has been used to design advanced systems. One such design is displayed in Fig. 11. The system incorporates the measured improved performance of the narrow waveguide channel width (1.0x2.25 mm) and catalytic gold electrodes, discussed above. The 4.5 m gain length is folded into 9 sections, milled into a single BeO ceramic slab. The schematic layout shown in Fig.11 has a custom advanced acousto-optic tuner attached. According to the model predictions for the system, operating on the $^{13}\text{C}^{18}\text{O}_2$ isotope for increased atmospheric transmission³, this laser can produce an average power of 30W (300 μJ / pulse) in a 44 line sequence at 100 kHz pulse repetition rate.

Fig. 11. Schematic drawing of a proposed laser system. Laser consists of 9- pass waveguide gain cell with a A/O tuner unit for rapid tuning.

VI. Future System Concepts

Whereas the CO_2 laser transmitter technology that has been developed thus far is robust and flexible, it would benefit from increased wavelength coverage¹³. We propose to expand coverage by employing nonlinear crystal technology to generate efficient tunable infrared beams by difference frequency generation (DFG). The approach has only recently become more feasible with reports¹⁴ of improvements in the transmission and power-handling capabilities of the infrared nonlinear crystal cadmium germanium arsenide (CGA). CGA has an extremely high figure of merit, but absorptions in the 4-12 μm wavelength range exceeding 0.4 cm^{-1} have restricted its use to low power applications. Recent reports of absorptions less than 0.1 cm^{-1} , achieved by irradiating the crystal with fast electrons, make it more likely that the crystal can endure the high average power requirements of remote sensing applications.

A. Difference frequency generation (DFG)

For difference frequency generation in the 8-12 μm atmospheric transmission band, an intense pump laser at a wavelength near 5 μm is mixed with a seed laser with a wavelength near 10 μm . With appropriate phase matching and sufficient power density to produce strong nonlinear conversion, the 5 μm pump is efficiently converted to high power at the two longer wavelengths corresponding to the seed frequency and difference frequency beams. Either the 5 μm pump, the 10 μm seed, or both can be tuned to produce tunable output at the difference frequency. A schematic diagram of a potential system using CO_2 lasers to generate both the pump beam and the seed laser beam is shown in Fig. 12. Tunable 5 μm light is first generated by frequency doubling a tunable 10 μm CO_2 laser, using the same type of nonlinear crystal as is used for difference frequency generation in the subsequent stage.

Fig. 12. Schematic diagram of experimental hardware configuration for generating tunable output (ω_5) by difference frequency generation (DFG) in a nonlinear crystal at frequency $\omega_5 = \omega_3 - \omega_4$, with ω_4 generated by a tunable CO_2 laser, and ω_3 generated in a separate nonlinear crystal by second harmonic generation (SHG) of a second tunable CO_2 laser.

Since the original pump light is tunable over about 200 cm^{-1} (see Fig. 13) the $5\text{ }\mu\text{m}$ light tunes over about 400 cm^{-1} , and mixing the latter with a tunable seed beam gives difference frequency light tunable over the entire 450 cm^{-1} atmospheric transmission band from 8 to $12.5\text{ }\mu\text{m}$. Model results of this DFG process are shown in Fig. 14. Since each CO_2 laser is tunable over as many as 60 laser lines, the output difference frequency obtained by mixing any pump line with any seed line, is tunable over $\sim 60 \times 60 = 3600$ lines, at intervals averaging less than 0.2 cm^{-1} . This constitutes a substantial improvement in both wavelength range and line spacing compared to the coverage presently available (Fig. 13) employing CO_2 lasers directly. While it is possible that solid-state lasers might eventually replace CO_2 lasers as pump sources for the difference frequency generation in this application, the rapid tunability and high repetition rates that we have demonstrated with CO_2 lasers gives them a major advantage for near-term demonstrations of the proposed approach in fieldable systems. Furthermore the high power and efficiency of CO_2 lasers, combined with the advantage of an order of magnitude smaller quantum defect than when starting from $1\text{ }\mu\text{m}$ solid state lasers, gives CO_2 lasers important advantages as pump sources for nonlinear generation of long wave infrared radiation.

Fig. 13. Standard CO_2 laser system and spectral coverage (Number of lines $N \sim 40$ -60). The strongest lines of the four bands in the CO_2 laser output give fair coverage of the spectral region from 9 - $11\text{ }\mu\text{m}$, but there are substantial gaps between the four bands, and the regions 8 - $9\text{ }\mu\text{m}$ and 11 - $12.5\text{ }\mu\text{m}$ are not covered.

Fig. 14. Difference frequency generation (DFG) with two CO_2 lasers by the arrangement shown in Fig. 12 permits coverage of the entire spectral range from 8 - $12.5\text{ }\mu\text{m}$ with 2000 - 3000 closely spaced lines. Different colors indicate bands from Fig. 13 that were frequency doubled to obtain $5\text{ }\mu\text{m}$ pump used in generating DFG.

B. Continuously tunable laser transmitters.

It may be possible to extend the approach outlined above, for generating *discrete* difference frequencies in a nonlinear crystal, to *continuous* tuning by replacing the tunable pulsed narrow-band CO_2 seed laser with a quantum cascade laser (QCL) having continuous tunability over a gain bandwidth the order of 20 - 50 cm^{-1} . Spanning the entire 8 - $12\text{ }\mu\text{m}$ atmospheric transmission band may require several QCL seed lasers, spaced at broad intervals throughout the band, in conjunction with the rapidly the tunable CO_2 pump laser.

Whether a CO_2 laser or a QCL supplies the seed wavelength, the gain for the nonlinear conversion must be supplied by an intense $5\text{ }\mu\text{m}$ laser pump pulse, and it is the energy in the pump pulse that is converted to

LWIR photons. The gain needs to be somewhat higher for the QCL seed than for conversion of the stronger CO₂ laser pulse. Since gain is exponential in pump pulse power, then assuming a gain of 10 is required to efficiently convert the pump pulse with a moderately intense CO₂ seed, and a gain of 10⁶ is required for a low power QCL seed, then the ratio of required pump powers would be about 6. This is not negligible, because efficient nonlinear conversion processes typically operate close to damage thresholds, so that even factors of two may be important. But in general, DFG should require less gain than unseeded optical parametric oscillators, which need to start up from only a few photons of spontaneous emission. DFG also provides better control of output wavelength and bandwidth.

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