Optics & Laser Technology 88 (2017) 263–274

Contents lists available at ScienceDirect

Optics & Laser Technology

journal homepage: www.elsevier.com/locate/optlastec

Full length article

Tunable CO_2 laser system with subnanosecond-pulse-train output

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ARTICLE INFO

Keywords:
CO_2 laser
Optical free induction decay
Subnanosecond

ABSTRACT

A CO_2 laser system has been demonstrated that generates a train of subnanosecond pulses tunable over the *P* and *R* branches of the CO_2 laser spectrum at 9–11 μm. It utilizes optical free induction decay to generate a single ~100-ps laser pulse from a tunable transverse-excited-atmospheric CO_2 laser. This laser pulse is injection-seeded into a high-pressure CO_2 oscillator whose output consists of a train of amplified ~100-ps pulses with maximum pulse energy of 30 mJ, corresponding to a peak power of 300 MW. The ~100-ps, tunable, infrared laser pulses are needed for a new technique to remotely detect atmospheric gaseous molecules, which relies on the train of CO_2 laser pulses selectively exciting the target molecules whose presence is then revealed using a separate terahertz probe beam.

1. Introduction

There is keen interest in developing ultrashort-pulse carbon dioxide (CO_2) lasers for applications such as laser-driven electron [1] and proton [2] acceleration, and Compton scattering *x*-ray sources [3]. Indeed, pulse lengths as short as a few picoseconds have been demonstrated [4,5]. However, as explained below, certain applications require of order 100-ps pulse lengths that are tunable over all the CO_2 laser lines.

Remote detection of gaseous chemical agents can be important for military and anti-terrorism applications. While discrete devices are available for sensing these agents, the widespread field deployment of these devices is impractical, especially during military operations. A laser system utilizing differential absorption lidar (DIAL) can be used to remotely detect these agents and is attractive because the laser beam can be swept 360° around the perimeter and from the horizon to the zenith. However, DIAL suffers from a fundamental limitation. It relies on detecting the absorption of the laser light tuned to the resonance of the target molecule. Atmospheric pressure broadening causes the absorption spectrum of the target species to overlap with the spectrum of other gases, thereby interfering with the unambiguous identification of the agents. This greatly diminishes the effectiveness of DIAL for this application.

This problem can be overcome by utilizing a novel double-resonance technique [6] in which a CO_2 laser beam excites the molecules of interest at a remote distance. A separate THz beam is sent to probe these excited molecules and results in emission at specific THz frequencies unique to each molecule. The modulated signal can be detected via background retro-reflection or backscattered light using highly sensitive THz detectors. This technique provides a sensitive method for uniquely identifying the molecules via a 3-D specificity matrix whose parameters consist of the CO_2 laser frequency, the THz probe frequency, and the time-resolved THz signal relaxation characteristics [6].

This double-resonance remote sensing (DRRS) technique establishes a new paradigm where the CO_2 laser beam only excites the molecules of interest rather than relies on resonant absorption. Consequently, atmospheric broadening actually helps the process by making it easier to excite the molecules off-resonance. This innovation can help expand the usage of lidar systems for atmospheric detection of trace gases.

DRRS relies on the rapid molecular collisional relaxation time, which at atmospheric pressure and temperature is of order 100 ps. Therefore, an ultrashort, ~100-ps laser pulse is needed so that this excitation pulse together with the rapid collisional relaxation can modulate the molecular THz emission or absorption on a time scale much faster than the ~1 s temporal atmospheric fluctuations. This is important for separating the molecular signal from signals due to clutter. The laser output wavelength also needs to be tunable over the 9–11 μm region in order to resonantly excite the molecules of interest. A pulse energy ≥10 mJ is needed for sensing at ranges of order 100 m [6]. Higher pulse energy will allow extending the sensing range. A pulse train is also highly desirable because it permits averaging over many pulses, thereby improving the signal-to-noise ratio (SNR). This will help increase the sensing range and enable more rapid scanning of air regions. Ultimately, the laser system must be portable and durable enough for deployment in the field. This implies the need to keep the CO_2 laser system as simple as possible in its design and operation.

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http://dx.doi.org/10.1016/j.optlastec.2016.09.022
Received 1 June 2016; Received in revised form 22 August 2016; Accepted 14 September 2016

This paper describes a tunable CO_2 laser system that generates a train of pulses with pulse lengths ~100 ps and pulses energies > 10 mJ. This system was designed and constructed to satisfy the needs for DRRS. As explained next, while various methods are possible for generating ultrashort CO_2 laser pulses, what distinguishes this work is its wavelength tunability and production of a train of pulses.

2. Design theory

For the DRRS application, pulsed transverse-excited-atmospheric (TEA) CO_2 lasers are the logical choice due to their high pulse energy, relatively compact design, well-established technology, and commercial availability. However, TEA CO_2 lasers typically have output pulse lengths of order 50–100 ns in a gain-switched spike. Various methods are possible for shortening the pulse length.

Q-switching of CO_2 lasers is possible [7–9]. Although high pulse energies are possible, the pulse lengths cannot be reduced to the subnanosecond level.

Both passive [10] and active mode-locking [11] have been used to generate short pulses. Passive mode-locking is generally more effective than active mode-locking at producing the shortest pulse lengths [12] with durations as short as 150 ps demonstrated [11]. However, passive mode-locking is also more difficult to control, especially if the individual pulses must be temporally synchronized with other pulses, such as with the THz probe for DRRS. A more serious issue is that DRRS requires the ability to readily tune to different wavelengths. Reestablishing mode-locking every time the wavelength is changed would be very difficult to do, especially in a field-deployed system.

Optical semiconductor switching [13], in which the CO_2 laser pulse passes through a semiconductor plate oriented at Brewster's angle to truncate the laser pulse, is an effective means for producing a shorter pulse with a controlled pulse length. A separate, fast-risetime, near-infrared (NIR) laser pulse illuminates the plate and creates free electrons (carriers) on the plate surface. These carriers act to reflect the CO_2 light, thereby truncating the CO_2 laser pulse length. Two plates are typically used to slice the front and rear of the laser pulse [14–16]. The minimum length of the sliced pulse is only limited by the risetime rate or, equivalently, the minimum pulse duration of the NIR laser beam illuminating the semiconductor slab. Sliced pulse lengths as short as 130 fs have been demonstrated [17]. A similar gating technique to slice the CO_2 pulse utilizes optical Kerr switching where a fast NIR laser pulse is used to switch the Kerr cell [18].

One drawback of semiconductor and optical Kerr switching is it requires a separate, ultrashort-pulse NIR laser to illuminate the semiconductor slabs or Kerr cell. This considerably complicates the system, which is highly unattractive for DRRS due to its adverse impact on system size, reliability, and cost.

More recently, chirped-pulse-amplification (CPA) has been applied to produce ultrafast IR seed pulses [5], in which a solid-state laser system featuring an optical parametric amplifier (OPA) is used to generate a ps seed IR pulse that is subsequently amplified by a CO_2 laser amplifier. Commercial solid-state OPO systems are available that are tunable in the IR region [19]; however, the pulse lengths are either in the nanosecond or picosecond ranges. There tends to be a gap in the ~100 ps pulse length range for solid-state lasers due to the inherent characteristics of the methods used to produce the pulses, i.e., Q-switching or mode-locking.

Quantum cascade lasers (QCL) are able to generate tunable radiation over 3–15 μm [20]. However, these are not viable seed sources for CO_2 lasers. The shortest pulse lengths produced by QCLs are typically 10′s ns [21] with pulse energies of order nJ, which is too low for effective injection seeding. Although progress is being made to produce ps pulses from QCLs [22], it is likely the eventual pulse energies will be again too low and, furthermore, the high-frequency output pulse train from these mode-locked lasers may be incompatible with seeding CO_2 laser amplifiers. Typically, a single subnanosecond pulse is needed as the injection seed and the seed should be generated at the same repetition rate as the CO_2 laser amplifier.

We should emphasize that although some of the preceding techniques are able to produce 100-ps laser pulses, they are often not easily tunable over the CO_2 lasing spectrum. For example, the semiconductor plates used during pulse slicing are oriented at Brewster's angle, which means this angle would need to be adjusted at each lasing wavelength due to the change in the index of refraction as a function of wavelength. Furthermore, this angle change would displace the beam, thereby requiring a realignment of the optical system. Thus, while wavelength-tuning of CO_2 lasers is normally very straightforward using an intracavity grating, typical methods for generating ultrashort CO_2 laser pulses are not necessarily compatible with wavelength tuning. As we shall explain next, our approach distinguishes itself by not only generating a train of 100-ps laser pulses, but also being easily tunable over the CO_2 lasing spectrum.

The technique we chose for generating the ~100-ps pulses is based upon a process called optical free induction decay (OFID) [23], in which the 100-ns CO_2 laser pulse is sent through a plasma shutter and then through a hot cell (400–500 °C) filled with CO_2 gas. The plasma shutter truncates the CO_2 laser pulse, but unlike in semiconductor switching, this truncation is not intended to shorten the CO_2 laser pulse. Instead the truncation generates high-frequency sidebands whose light also enters the hot cell. The purpose of the hot cell is to act as a frequency filter, whereby it selectively absorbs light at the fundamental frequency, but allows the sideband light to pass through [24]. The frequency spectrum of the light exiting the hot cell is therefore much broader than the incoming pulse's frequency bandwidth and, hence, corresponds to pulses with much shorter lengths. OFID-generated pulses as short as 30 ps have been demonstrated [25]. By adjusting the pressure in the hot cell, which essentially changes its filtering characteristics, it is possible to obtain output pulse lengths between 30 and 300 ps [26].

Thus, the OFID technique is able to provide ~100 ps CO_2 laser pulse lengths in an essentially passive manner that are easily wavelength tunable. This makes it less complicated than semiconductor/Kerr cell switching and, consequently, a suitable candidate for DRRS.

After exiting the hot cell, the ~100-ps pulse has a typical energy of order 10′s μJ. A CO_2 laser amplifier is needed to amplify the pulse to useful energy levels. CO_2 lasers rely on vibrational-rotational molecular transitions for the laser emissions, which means the gain profile consists of discrete frequencies separated by ~40–55 GHz (~1.3–1.5 cm^{-1}). This allows the laser to generate discretely tunable radiation over 9–11 μm, but the discrete gain profile also complicates the ability to amplify ultrashort laser pulses because the gain bandwidth is not continuous over the wide bandwidth of the ultrashort pulses. Hence, the CO_2 discrete gain spectrum must be broaden out to amplify the ~100 ps pulse. The easiest way to accomplish this is to operate the CO_2 amplifier at high pressure (e.g., ~10 atm) so that pressure broadening (~5 MHz/Torr) causes the gain line spectra to widen and merge with each other [27]. This pressure broadening of the CO_2 gain spectrum is illustrated in Fig. 1, where Fig. 1(a) shows the discrete gain lines at 1 atm and Fig. 1(b) depicts the smearing together of the gain lines at 10 atm.

An alternative method for broadening the CO_2 laser gain spectrum is to use gas mixtures containing different isotopes of CO_2 [28]. The gain lines are slightly shifted for each isotopic molecule. Hence, the gain profile of a mixture of isotopes contains many more gain lines in between the nominal ones. This helps smooth out the gain profile with pressure broadening still used to blend the gain lines together. The advantage of using isotopic gas mixtures is it can require less gas pressure to smooth out the gain profile. The disadvantage of isotopic gas mixtures is their high expense.

Utilizing strong field broadening of the lasing transition is another means for amplifying picosecond IR pulses that can be done at atmospheric pressures [29]. This was demonstrated using 3-ps pulses.