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Ultra-short pulse generation with CO_2 lasers

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Although considerable progress has been made, and several methods now permit the generation of 10 μm pulses in the picosecond range, mode-locked CO_2 oscillators do not appear to have reached their full potential. Two techniques, passive and injection mode-locking, have been used to generate pulses shorter than 100 ps and of these injection mode-locking seems to be particularly well suited to CO_2 oscillators since it is less sensitive to the short gain lifetime and rapid gain risetime of multi-atmosphere discharges. To date, no attempt to use both techniques simultaneously has been reported for CO_2 although this approach has been applied successfully to Nd:YAG oscillators (Murray & Lowdermilk 1978).

Injection mode-locking is clearly a valuable tool for the study of pulse broadening mechanisms as a function of the operating pressure of the slave laser and the intensity, duration and wavelength of the injected pulses. Such investigations are of fundamental importance to the further development of ultra-short pulse CO_2 oscillators and amplifiers, and may well lead to the routine generation of pulses as short as 10 ps.

Introduction

The generation of 10 μm pulses with durations of 1 ns or less has become increasingly important in recent years. Much of the motivation for producing such pulses has originated in the area of laser fusion research; however, ultra-short pulses of mid-infrared radiation also have significant potential in a number of other areas such as photochemistry, isotope separation, dynamic spectroscopy, semiconductor physics and laser ranging.

Depending on the requirements of a particular application, various techniques are now available for the generation of pulses with durations ranging from a few nanoseconds down to *ca.* 10 ps. Two approaches that are particularly interesting are optical free induction decay (Yablonovitch & Goldhar 1974) and semiconductor switching (Alcock *et al.* 1975*a*), both of which permit the gating of very short pulses from a conventional gain switched CO_2 laser pulse. However, the pulses generated with these techniques are limited to relatively low energies. It is therefore natural to exploit the mode-locked oscillator approach wherever possible, since a significant fraction of the stored energy is available in an individual pulse. Unfortunately, the conventional mode-locking techniques, which have been applied so successfully to many visible and near infrared lasers, are less well suited to CO_2 oscillators. As a result, somewhat different approaches are being pursued to optimize the generation of mode-locked pulses in the picosecond range.

A major factor limiting the success of conventional mode-locking techniques is the limited bandwidth of long pulse or continuous wave (c.w.) CO_2 lasers and the short gain lifetime of systems having the bandwidth required for picosecond pulse generation. Nevertheless long pulse and c.w. operation of near atmospheric pressure CO_2 lasers has been reported (Wood *et al.* 1975; Matsushima *et al.* 1978) and such devices, with a bandwidth of *ca.* 3 GHz, could provide pulses as short as *ca.* 150 ps. Much higher pressure operation is possible in pulsed lasers using self-sustained transverse discharges (Hidson *et al.* 1972; Alcock *et al.* 1973*a*), E-beam sustained discharges (Basov *et al.* 1973*a*, *b*) or optical pumping (Chang & Wood 1973) with an appropriate source of pulsed infrared radiation. The principal advantage of increased pressure operation was first pointed out in 1971 by Basov *et al.* who noted the large available bandwidth provided by the overlapping of vibrational–rotational lines that occurs at pressures significantly higher than 1 atm† (figure 1) (Taylor *et al.* 1979). However, the increased pressure introduces other less desirable effects, since both the excitation and de-excitation of the active laser levels are dominated by collisional processes. Thus both the gain risetime (figure 2) (Alcock *et al.* 1975*b*) and lifetime (Basov *et al.* 1973*b*) decrease with increasing pressure and, as will be shown below, this significantly influences the effectiveness of standard mode-locking techniques.

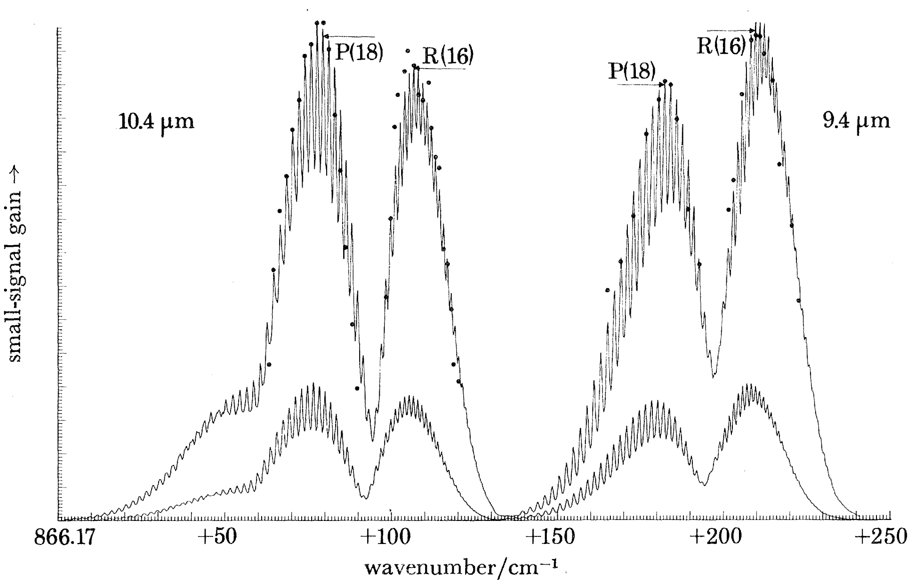

FIGURE 1. Plot of experimental data points and theoretical gain prediction as a function of CO_2 frequency for a laser pressure of 10 atm and a pump energy density of 110 J l^{-1} atm^{-1}. The theoretical prediction is normalized to the experimental gain on the P(18) 10.4 μm line. The bottom curve represents the contribution of sequence bands to the gain.

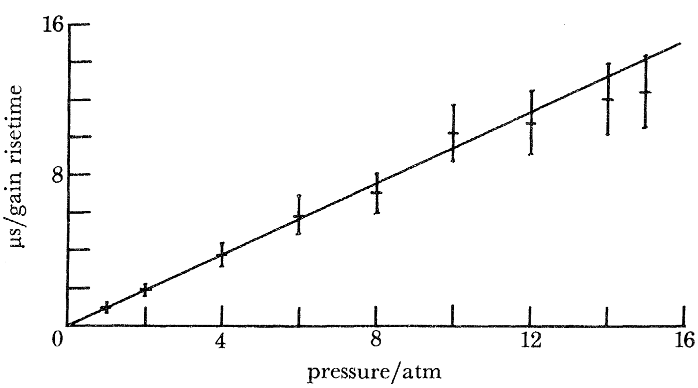

FIGURE 2. Measured inverse gain risetime plotted as a function of pressure for a u.v. preionized TE CO_2 discharge.

† 1 atm ≈ 10^5 Pa.

Passive mode-locking

The use of saturable absorbers to passively mode-lock many solid state and dye lasers is now a well proven, reliable technique capable of yielding reproducible pulse trains with individual pulses having a typical duration of a few picoseconds.

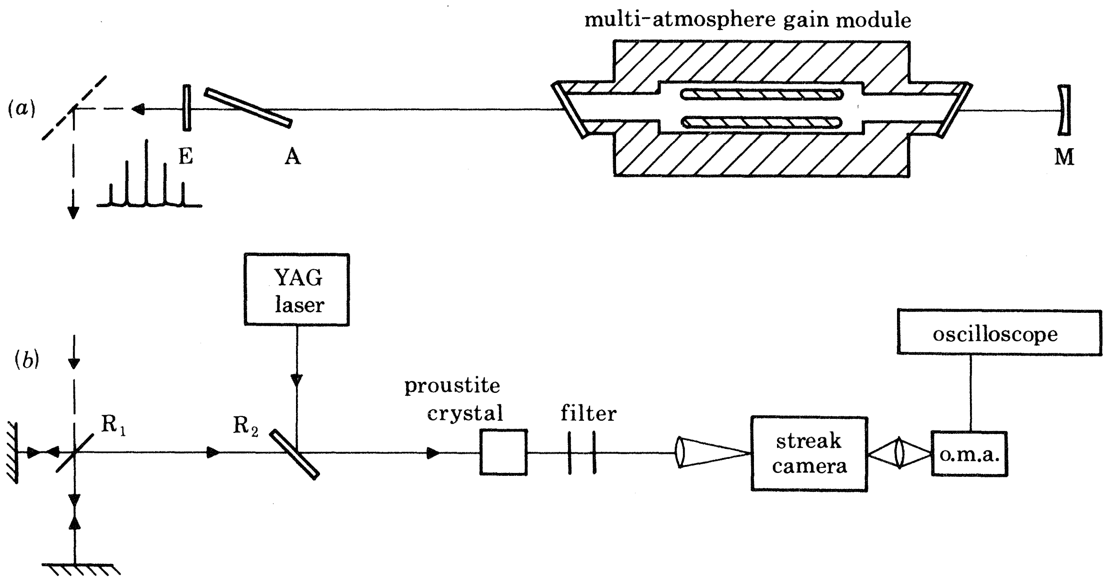

Figure 3. Schematic illustration of (*a*) a passive mode-locking of a TE CO_2 oscillator with p-type germanium, and (*b*) the up-convertor detection system used to resolve 10 μm pulses of duration greater than about 40 ps.

In early experiments with TEA CO_2 lasers a similar approach was adopted, and the generation of mode-locked pulse trains was reported by several authors who used molecular gases (Nurmikko *et al.* 1971; Fortin *et al.* 1973; Lavigne *et al.* 1975), hot CO_2 (Gibson *et al.* 1971) or p-type germanium (Gibson *et al.* 1974) as the saturable absorber medium. From the point of view of short pulse generation, the *ca.* 3 ps relaxation time and broad bandwidth of p-type germanium (Keilmann 1976) are particularly attractive, and the use of this material in a multi-atmosphere CO_2 laser has yielded the shortest 10 μm pulses (*ca.* 80 ps) produced by passive mode-locking (Alcock & Walker 1974; Walker & Alcock 1974). In that experiment (figure 3*a*) two thin slabs of p-type germanium were inserted in the laser resonator, one at the Brewster angle, the other an etalon (essential for stable mode-locking) at normal incidence where it acted as both output coupler and mode-locking element. This configuration resulted in the pulse train shown in figure 4*a* when the high pressure CO_2 discharge was operated at 12 atm.

Measurement of the duration of ultra-short 10 μm pulses requires special techniques since electro-optical streak cameras (the most versatile real-time recording systems for picosecond visible and near i.r. pulses) are not directly applicable at wavelengths substantially longer than 1 μm. For the real-time detection of ultra-short CO_2 pulses it is therefore necessary to transfer the temporal characteristics of the 10 μm pulses to a shorter wavelength where photocathodes and thus conventional streak cameras do respond. One technique, mixing with shorter wavelength radiation (1.06 μm) in proustite to generate the sum frequency (0.96 μm) has the advantages of broad bandwidth (Walker & Alcock 1976) and linearity over a wide dynamic range (Jaanimagi *et al.* 1979).

In order to measure the duration of the passively mode-locked pulses shown in figure 4*a* such an up-conversion system with an overall temporal resolution of 40 ps was employed (figure 3*b*). This permitted the detection of mode-locked pulses as short as 80 ps (figure 4*b*) and even shorter spikes in the non-mode-locked output of the multi-atmosphere CO_2 oscillator (figure 4*c*).

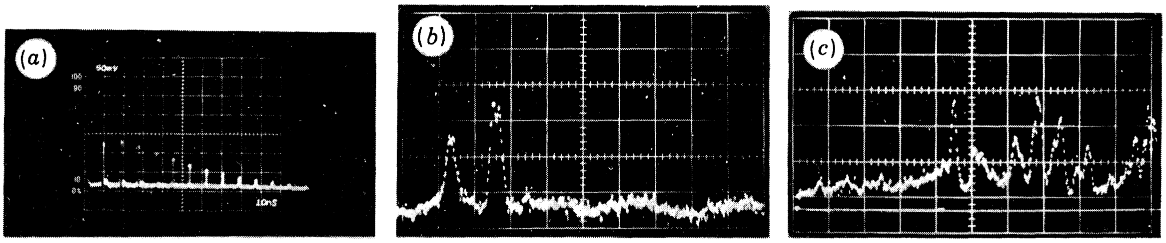

FIGURE 4. Oscilloscope traces of the passively mode-locked output of a 13 atm TE CO_2 oscillator: (*a*) portion of a pulse train detected directly by a photon drag detector and Tektronix 7904 oscilloscope (10 ns per division); (*b*) up-converted pulses from the mode-locked CO_2 laser (200 ps per division); (*c*) non-mode-locked output of the TE CO_2 oscillator (200 ps/per division).

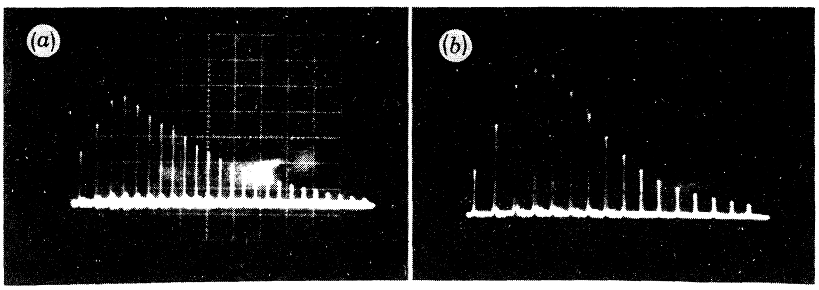

FIGURE 5. Oscilloscope traces illustrating active and injection mode-locking: (*a*) train from actively mode-locked TE CO_2 oscillator used as the master oscillator in (*b*) (pulse separation 13.3 ns); (*b*) output train from injection mode-locked slave laser operated at a pressure of 4 atm (pulse separation 13.3 ns).

Although it is possible that shorter pulses could be generated by passive mode-locking, the very rapid build-up of the gain and the fairly low damage threshold of the germanium elements do not facilitate generation of highly reproducible pulse trains. As a result, passive mode-locking with germanium has not been widely applied although it is undoubtedly the simplest method for the production of subnanosecond CO_2 pulses and its full potential may not have been realized.

ACTIVE MODE-LOCKING

The use of intra-cavity modulators to actively mode-lock TEA CO_2 lasers has resulted in the generation of pulses with a duration of *ca.* 1 ns (Wood *et al.* 1970; Abrams & Wood 1971). This approach offers highly reliable operation of mode-locked atmospheric pressure oscillators and has the significant advantage that the mode-locked pulses are synchronized with the electronic oscillator used to drive the modulator.

With the development of multi-atmosphere TE CO_2 discharges, active mode-locking was also investigated (Alcock & Walker 1974). However, very little pulse shortening was observed

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as the pressure of the active medium and hence its bandwidth was increased. Figure 5*a* shows the pulse train generated when a Brewster angle modulator, driven at 40 MHz, was inserted in the resonator of a 12 atm transverse discharge device. Although clean trains of nanosecond mode-locked pulses were consistently observed with a nanosecond response detection system, no significant reduction in the recorded pulse duration was observed when the laser output was up-converted and displayed on a 1 GHz bandwidth oscilloscope.

Since both the gain risetime and gain lifetime are reduced in proportion to the operating pressure, the weak dependence of the pulse duration on the amplifying bandwidth can be understood in terms of the transient mode-locking analysis of Kuizenga *et al.* (1973). They show that after a sufficient number of cavity transits for the pulse shape to become essentially Gaussian, the pulsewidth can be approximated by

$$\tau_p = \sqrt{\left(\tfrac{1}{2}\ln 2\right)}\,\frac{1}{\pi}\frac{1}{\theta_m}\frac{1}{\sqrt{M f_m}}, \tag{1}$$

provided that τ_p is still long in comparison to the steady-state pulse duration. In (1), θ_m is the depth of modulation, f_m is the modulation frequency and M is the number of cavity round-trips. For the 12 atm laser described above, the gain lifetime of *ca.* 1 μs will permit less than 100 round-trips for a modulation frequency of 80 MHz. Assuming a depth of modulation of 0.4, substitution in (1) yields a pulse duration of *ca.* 0.6 ns, which is close to the experimentally measured values. Thus, although some decrease in pulse duration could be achieved by increasing the depth of modulation and the modulation frequency, the main limitation is the relatively small number of cavity transits in comparison to the *ca.* 10^4 round-trips required to reach the steady-state pulse duration governed by the gain bandwidth. For the steady state régime to be reached, the gain lifetime in multi-atmosphere CO_2 devices would have to be increased to more than 100 μs. In principle this condition could be met, for example in an E-beam sustained device; however, such long pulse operation has not been reported.

Injection mode-locking

A technique that may well permit the bandwidth of multi-atmosphere CO_2 discharges to be fully exploited is the regenerative amplification or injection mode-locking scheme first reported by Belanger & Boivin in 1974. In this approach (figure 6), a very low intensity pulse is injected into the laser resonator during the gain build-up and is subsequently regeneratively amplified. With appropriate timing and control of the injected pulse amplitude, this can be made to occur before spontaneous emission is able to build up and generate the normal gain-switched output pulse. Under these conditions the output consists of a mode-locked pulse train containing virtually all of the available energy and a negligible contribution from spontaneous emission. Since it was first demonstrated, injection mode-locking has been applied to TE CO_2 lasers (Belanger & Boivin 1974, 1976; Corkum *et al.* 1977; Alcock *et al.* 1977*b*), dye lasers (Moses *et al.* 1976) and Nd:YAG oscillators (Murray & Lowdermilk 1978), and as a mode-locking technique offers several significant advantages. These include: generation of short pulses having a duration that is not strongly dependent on the gain build-up time; automatic synchronization of the mode-locked pulse train with an externally controlled signal; compatibility with many relatively simple pulse-forming techniques; absence of mode-locking elements in the laser resonator, thus reducing the probability of damage due to high intra-cavity flux.

Although injection mode-locking has been extensively studied with TEA CO_2 oscillators, an early experiment (figure 6*a*), carried out with a 1.3 cm aperture multi-atmosphere laser has greater significance for ultra-short pulse generation (Alcock *et al.* 1977*b*). In that experiment a train of *ca.* 1 ns duration pulses (figure 5*a*) from a small actively mode-locked, multi-atmosphere oscillator was used to control the output of a 70 cm^3 slave oscillator. This device comprised a 40 cm long discharge and a confocal unstable resonator formed by a 10 m radius of curvature concave mirror and a 6 m radius of curvature convex mirror. At an operating pressure of *ca.* 8 atm and with no injected radiation, this oscillator produced a 30 ns gain-switched pulse containing an energy of *ca.* 3 J. Successful mode-locking required careful matching of the

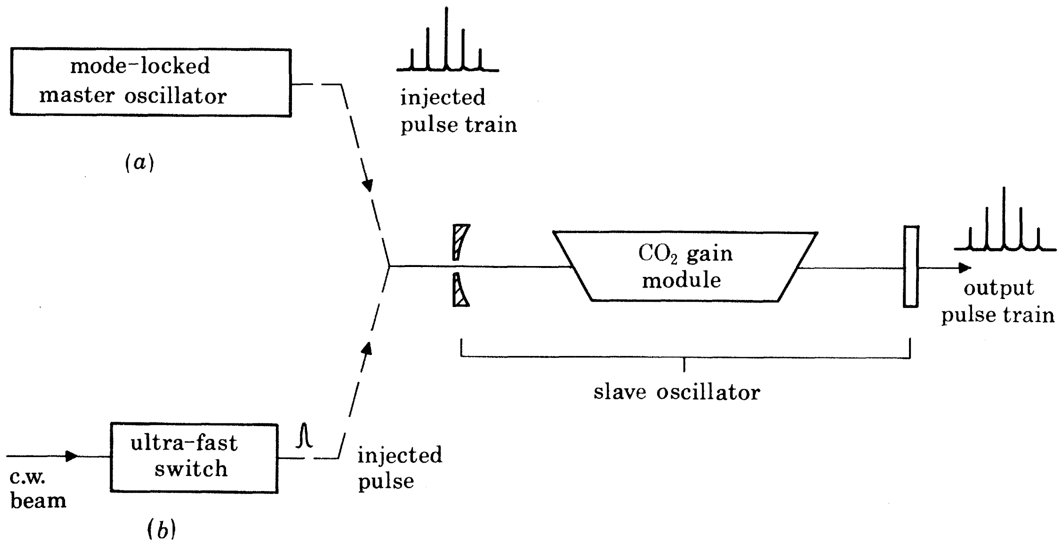

FIGURE 6. Schematic illustration of injection mode-locking with the use of either (*a*) a mode-locked master oscillator, or (*b*) a single 10 μm pulse gated from the output of a c.w. CO_2 oscillator.

'slave' and 'master' oscillator resonator lengths and appropriate timing of the two laser discharges. An example of the pulse train generated when the slave oscillator was operated at a pressure of 4 atm and with *ca.* 10 μJ of injected radiation is shown in figure 5*b*. Significantly shorter pulse trains were generated at higher pressures. At 8 atm a peak pulse energy of *ca.* 1 J was obtained in a pulse train containing a total energy of *ca.* 3 J. Although no detailed study of the individual pulse shapes or durations was attempted, both the input and output pulse trains were detector limited when monitored by a photon drag detector used in conjunction with a 500 MHz oscilloscope. The above results indicated the feasibility of applying to multi-atmosphere oscillators many of the improved injection mode-locking techniques developed initially with atmospheric pressure devices. In particular, experiments with TEA CO_2 lasers demonstrated that (*a*) mode-locking could be achieved by injecting a single pulse into the slave resonator at the appropriate time (Belanger & Boivin 1974, 1976) (figure 6*b*), and (*b*) pulses gated from the low power c.w. sources contain sufficient energy to effectively mode-lock the output of relatively large volume (*ca.* 5 l) transverse discharge lasers (Alcock *et al.* 1977*a*).

Injection mode-locking thus introduces the prospect of utilizing a broad range of short-pulse generation techniques that may not directly yield the high peak powers required for many applications. Such techniques include fast electro-optical switches (McLellan & Figueira

1978), optical free induction decay (Kwok & Yablonovitch 1977), stimulated Raman scattering in gases (Loree *et al.* 1976), and semiconductor switching (Alcock *et al.* 1975*a*; Alcock & Corkum 1979). The last three techniques are particularly interesting since they have permitted the generation of 10 μm pulses significantly shorter than 100 ps.

In addition, other nonlinear processes such as three-wave mixing, which have resulted in picosecond pulse generation at shorter wavelengths (Laubereau *et al.* 1978), should be applicable in the 10 μm region.

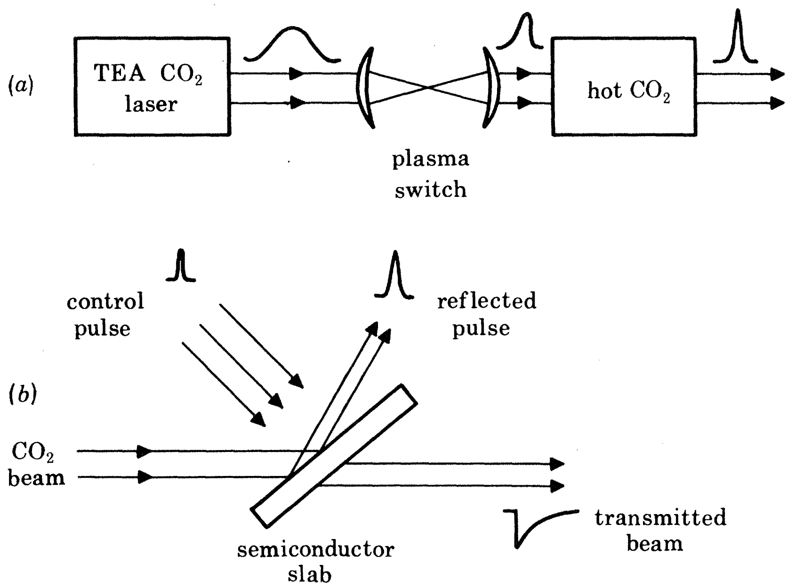

FIGURE 7. Schematic illustration of the two most common techniques for generating ultra-short 10 μm pulses: (*a*) free induction decay; (*b*) semiconductor switching.

Of the techniques capable of producing picosecond pulses, optical free induction decay and semiconductor switching have been most extensively studied. The former (figure 7*a*), developed by Yablonovitch & Goldhar (1974), exploits the rapid onset of attenuation due to the plasma formed in CO_2 laser-induced gas breakdown. The abrupt termination of a relatively long, single-mode TEA laser pulse is used in conjunction with a cell of hot CO_2 (which discriminates against the narrow bandwidth radiation) to generate a short 10 μm pulse by optical free induction decay. Pulses as short as 30 ps have been generated in this manner (Kwok & Yablonovitch 1977) and could clearly be used as a source for injection mode-locking.

The second approach, 'optical semiconductor switching' (figure 7*b*), involves the use of near infrared or shorter wavelength radiation to generate a high density of free carriers in a semiconductor (such as germanium) that is otherwise transparent to the 10 μm beam. As shown in figure 8, the Brewster angle reflectivity of germanium is a rapidly varying function of the free carrier pair density near the critical density of 1.6×10^{19} cm^{-3}. For short pulses of suitable wavelength control radiation (Alcock & Corkum 1979) such densities can be produced near the semiconductor surface at relatively low fluences. This is illustrated in figure 9 where the calculated surface plasma density in germanium is plotted as a function of time during and after illumination by a 10 ps pulse of either 0.6 μm (0.5 mJ/cm^2) or 1.06 μm (1 mJ/cm^2) control radiation.

Figure 9 also shows that the surface plasma density decreases rapidly after the 10 ps control pulse. In germanium this is almost entirely due to ambipolar diffusion of the free carriers into the bulk material (Auston & Shank 1974). However, in many semiconductors, recombination must also be considered. As the carrier pair density falls below the critical value, the reflectivity decreases rapidly (figure 8) thus terminating the reflexion switching process.

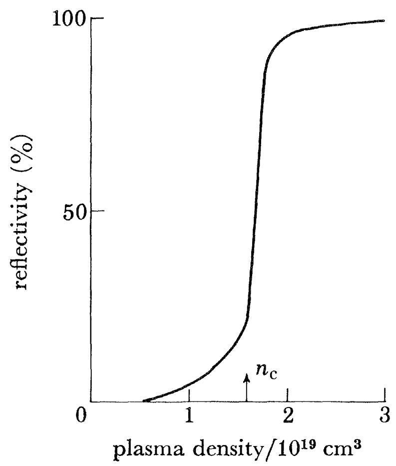

FIGURE 8. Calculated Brewster angle reflectivity plotted as a function of carrier pair density for 10.6 μm radiation incident on an optically thick germanium plasma.

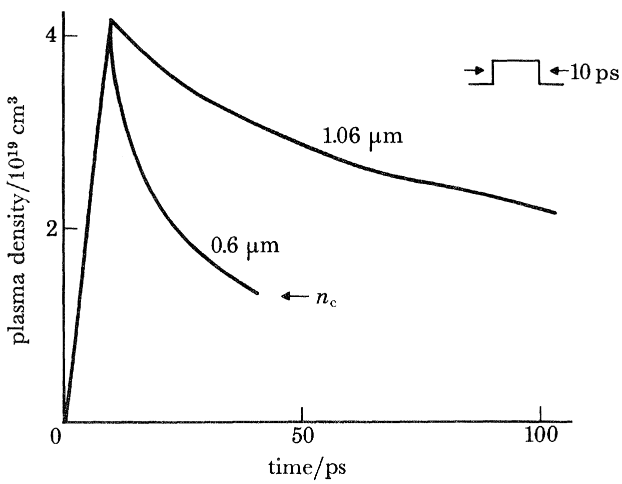

FIGURE 9. Surface density of free carriers in germanium plotted as a function of time for a 10 ps, 1 mJ/cm^2 (absorbed energy) 1.06 μm pulse (upper curve) and for a 10 ps, 0.5 mJ/cm^2 (absorbed energy) 0.6 μm pulse (lower curve).

The complementary process, optically controlled transmission through a semiconductor, is also illustrated in figure 7*b*. The 10 μm transmission is governed by both free carrier absorption and the modification of surface reflectivity described above. In the 10 μm region, free carrier absorption in germanium is dominated by an intravalence band transition having a cross section of 6×10^{-16} cm^2 (Gibson *et al.* 1972). (This is the same transition responsible for the saturable absorption properties of p-type germanium discussed previously.) The transmission can be abruptly terminated by free carrier absorption alone (figure 10) if the semiconductor is illuminated with a sufficiently intense control pulse. The recovery of the transmission is not plotted in figure 10 since it results from recombination, a process that requires many microseconds in typical germanium samples.

The shortest 10 μm pulses reported to date (figure 11) have been gated from the output of a single longitudinal mode TEA CO_2 laser (Jamison & Nurmikko 1978) using optical semiconductor switching. A germanium reflexion switch controlled by a *ca.* 10 ps, 1.06 μm pulse produced a rapidly rising 10 μm signal, which was then terminated by a second switch operated in the transmission mode. This technique is independent of both diffusion and recombination

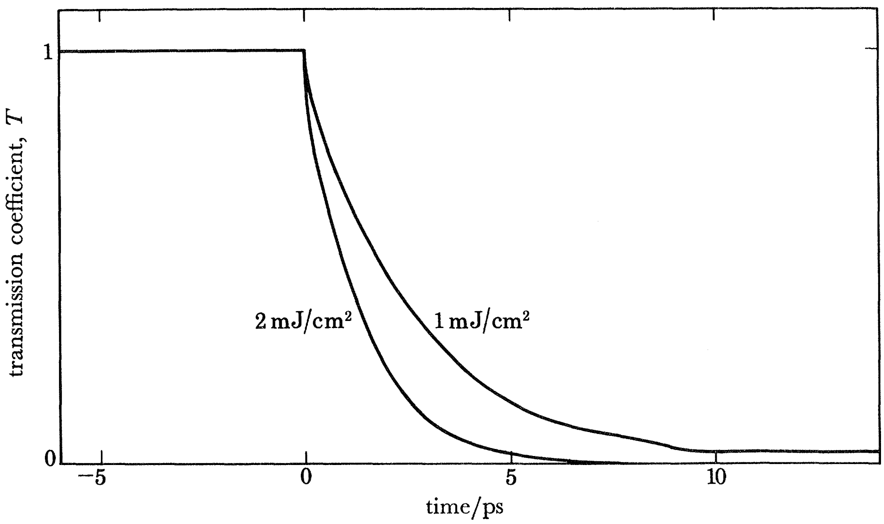

FIGURE 10. Attenuation of 10 μm radiation due to intravalence band transition in germanium under illumination by a 10 ps pulse of 1 μm radiation: upper curve, absorbed energy density = 1 mJ/cm²; lower curve, absorbed energy density = 2 mJ/cm².

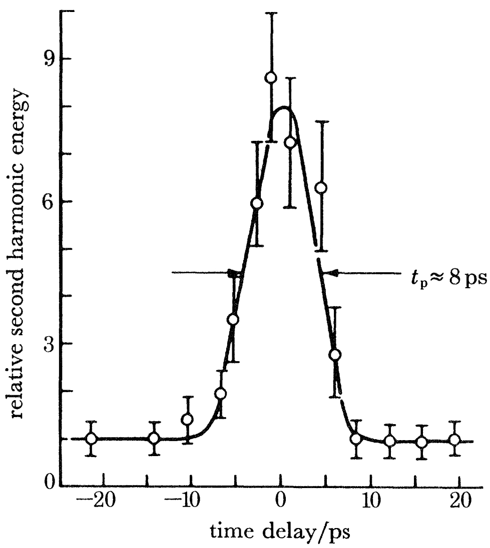

FIGURE 11. Autocorrelation measurement of the 10 μm pulse duration gated from the output of a single longitudinal mode TEA CO_2 oscillator with the use of optical semiconductor switching. (From Jamison & Nurmikko (1978).)

in the semiconductor and should therefore permit the generation of even shorter duration 10 μm pulses.

Semiconductor switching is the only technique that has been used to generate ultra-short pulses for injection mode-locking CO_2 oscillators. In one such experiment (Corkum *et al.* 1978) (figure 12), the output of a passively mode-locked flashlamp-pumped dye laser (figure 13*b*, lower trace) was directed onto a germanium slab at an energy density of *ca.* 0.5 mJ/cm^2 per mode-locked pulse. The switched radiation, consisting of a series of reflected pulses, was gated from the output of a *ca.* 1 W c.w. CO_2 laser and injected into a slave oscillator comprising a 26 cm × 0.7 cm × 0.7 cm 8 atm discharge and a stable resonator. The round-trip time of the

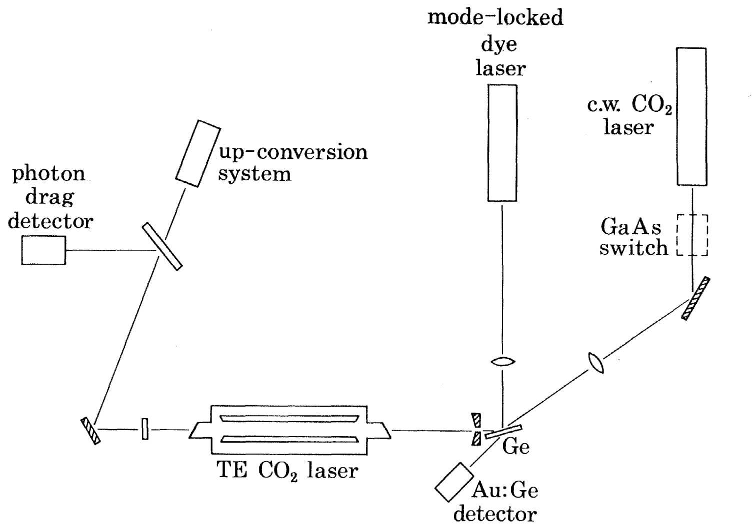

FIGURE 12. Experimental apparatus used to injection mode-lock a multi-atmosphere TE CO_2 oscillator and to measure the duration of the resulting mode-locked pulses.

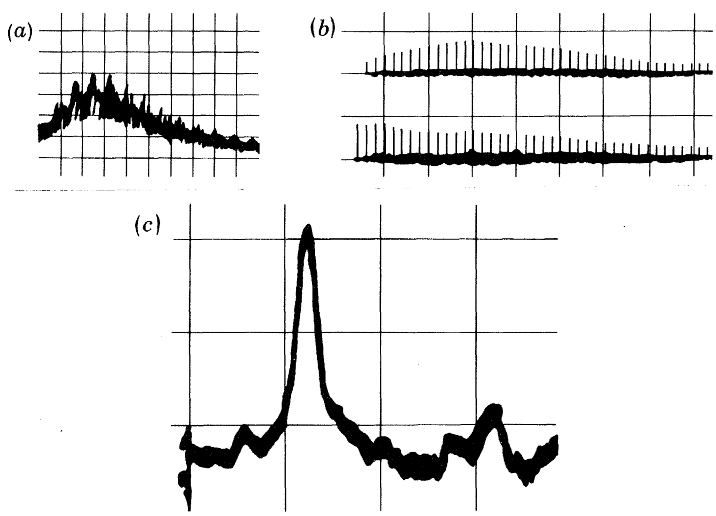

FIGURE 13. Oscilloscope traces illustrating the regenerative amplification of picosecond pulses gated with a semiconductor switch: (*a*) gain switched output of multi-atmosphere TE CO_2 oscillator with no injected radiation (20 ns per division); (*b*) mode-locked output of the CO_2 oscillator (upper trace) and synchronized train of 0.6 μm pulses (lower trace) (20 ns per division); (*c*) single high power pulse from the mode-locked output of the CO_2 oscillator operated at 4 atm (800 ps per division).

slave CO_2 laser resonator was accurately adjusted to be an integral multiple (4) of the dye laser cavity transit time (4 ns). With this arrangement it was not necessary to select a single dye or 10.6 μm pulse and, in addition, the initial synchronization between the 10.6 μm and 0.6 μm pulse trains was maintained.

Figure 13*a* shows the *ca.* 250 mJ output of the slave laser operated at 7 atm when no radiation was injected into the cavity, while figure 13*b* shows the injection mode-locked output (upper trace) with the 4 ns periodicity of the dye laser (lower trace). In this experiment, individual pulse durations were measured down to the 200 ps resolution limit of the detection system (figure 13*c*), while in another similar experiment pulses shorter than 100 ps were resolved (Corkum & Alcock 1978). Thus the applicability of injection mode-locking to multi-atmosphere CO_2 lasers has been demonstrated. When scaled to higher pressures this technique should permit the generation of significantly shorter pulses.

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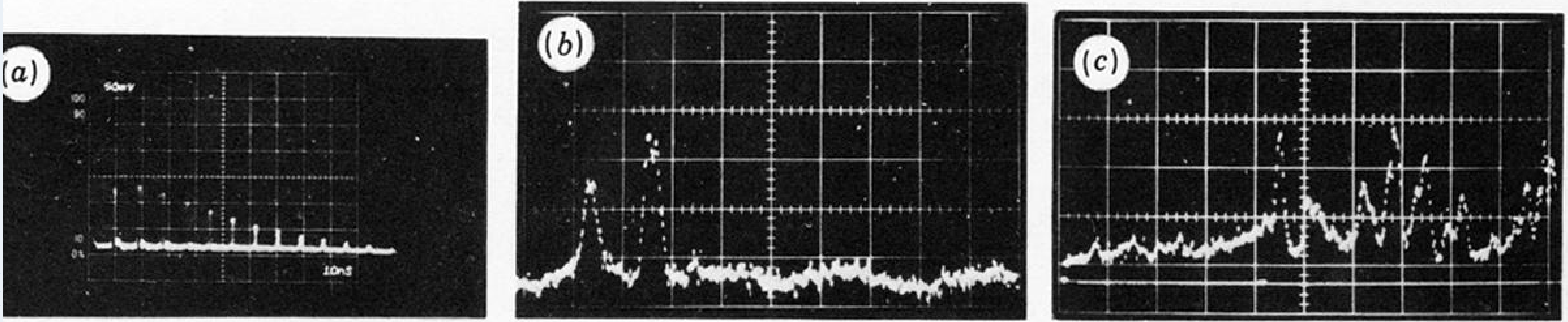

IGURE 4. Oscilloscope traces of the passively mode-locked output of a 13 atm TE CO_2 oscillator: (*a*) portion of a pulse train detected directly by a photon drag detector and Tektronix 7904 oscilloscope (10 ns per division); (*b*) up-converted pulses from the mode-locked CO_2 laser (200 ps per division); (*c*) non-mode-locked output of the TE CO_2 oscillator (200 ps/per division).

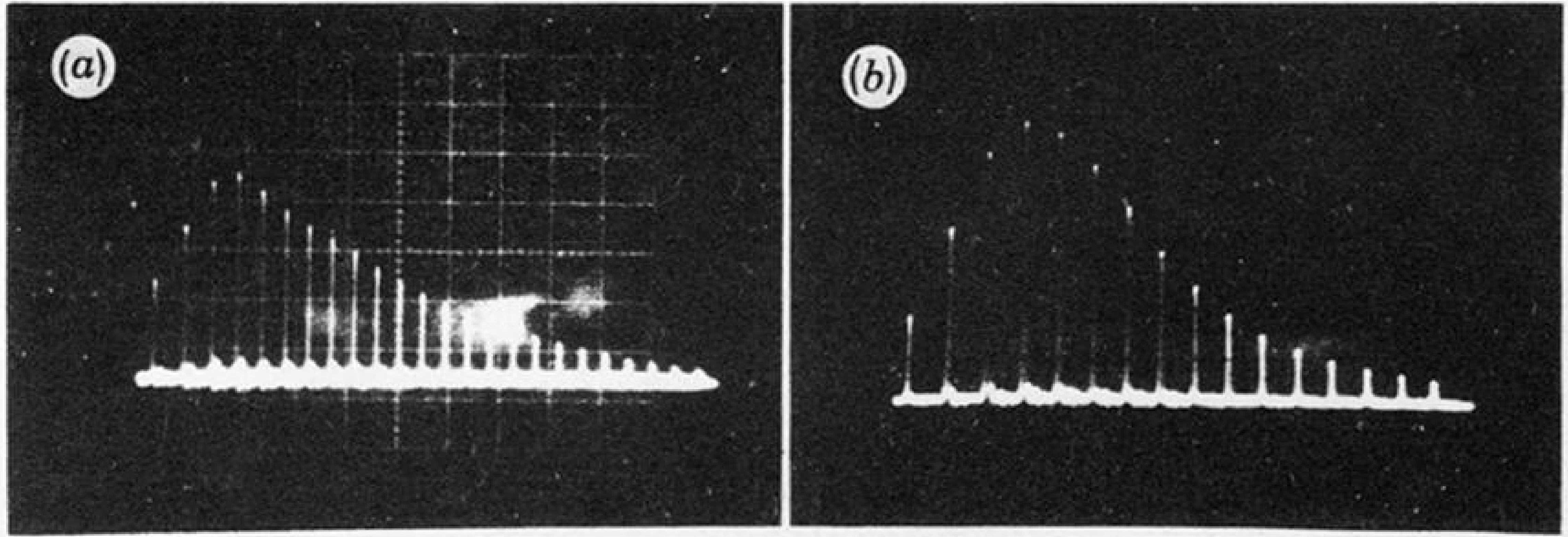

FIGURE 5. Oscilloscope traces illustrating active and injection mode-locking: (*a*) train from actively mode-locked TE CO_2 oscillator used as the master oscillator in (*b*) (pulse separation 13.3 ns); (*b*) output train from injection mode-locked slave laser operated at a pressure of 4 atm (pulse separation 13.3 ns).