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Multiwavelength Q-Switched CO_2 Laser with Continuous Discharge

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Abstract. A low-pressure (20 mbar) CO_2 laser allows to extract pulses at several selected wavelengths simultaneously from the same active medium. We demonstrated this, using an industrial laser modified by a Q-switch and a resonator with two branches. In one branch the wavelengths are spatially separated, whereas in the other they oscillate in one common transverse mode. We designed a multiwavelength resonator which requires a single additional reflector compared to usual laser cavities. It provided tunable oscillation at six wavelength simultaneously.

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There have been many attempts to operate pulsed CO_2 lasers on two or several of its vibrational-rotational transitions simultaneously or nearly simultaneously. A comprehensive compilation has been published recently [1, 2]. There have been several purposes of such work:
– The development of a light source for analytical measurements by the differential absorption method.
– The extraction of more energy from the laser medium. This is important for short-pulse lasers such as those used for laser fusion studies.
– The improvement of infrared multiphoton excitation of molecules, e.g., for isotope separation. This effect is based on the improved chances for resonances in multistep excitation, if more than one wavelength is offered to the molecule.

The latter purpose was our motivation. Multiwavelength excitation of $CHClF_2$ has been successfully applied to separate carbon isotopes [3–6]. Transversely excited atmospheric pressure (TEA) CO_2 lasers have been used in these experiments. On the other hand, we have demonstrated ^{13}C separation with this molecule, using a Q-switched laser with continuous discharge [7–9]. The advantage is that a (modified) industrial (highly reliable) CO_2 laser can be used, which can deliver photons at lower cost than TEA lasers [9].

Of course, two or more wavelengths can also be generated by separate lasers. However, in this approach it is difficult to guarantee a desired time delay of the pulses and the spatial overlap of the beams over long pathlengths (which is needed in isotope separation). Our design solves both problems and has the additional advantage that only one laser is needed [10].

Extraction of two wavelengths from the same active volume presents one problem: The delay and the relative energies of the two pulses will not be reproducible if they compete for the same inversion. Such a competition is induced by rotational relaxation and relaxation between the Fermi-coupled levels of CO_2. A radical solution for this problem is a spatial wavelength separation in the discharge region [5, 11–14]. This method, however, suffers from an inefficient use of the active medium (for geometrical reasons). Also orthogonal polarizations of the two oscillating wavelengths seems to ease the problem [15].

On the other side, line competition is certainly avoided if the pulses are short enough or if the laser gas pressure is low enough. (It is sufficient if the relaxation is not complete during the first half of the pulse.) The survey [1] confirms this general rule. For atmospheric pressure TEA lasers, this condition is met by mode-locked pulses (one to a few ns long). Although such short pulses would be desirable for isotope separation, it is not easy to generate them at high average power. However, in our laser, which is operated at 20 mbar, the condition for weak competition is already met for usual Q-switched pulses (200–300 ns). This is an important advantage of our laser for application in isotope separation.

1 Resonators for Multiwavelength Operation

Usually a wavelength-selective resonator consists of an end mirror and a grating or prism in autocollimation (Littrow)

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arrangement. The grating acts as a mirror for the desired wavelength. Some modifications of this configuration permit multiwavelength oscillation in the same active volume.

1.1 Resonator with a Low Dispersion Grating

A most simple, but not flexible way is to use a low dispersion grating. In our initial experiments we employed a grating with 30 grooves per mm [16]. Then the direction of oscillation for, e.g., the $9P20$ and $9P22$ lines differs by only 0.25 mrad, an angle which is 4 times smaller than the natural divergence of a diffraction limited mode with waist diameter $2w_0 = 6$ mm. With careful alignment of the grating angle, we obtained equal power on two adjacent lines, usually accompanied on each side by the next neighbor, which was about 10 times weaker.

1.2 Unsymmetric Binary Cavity

To achieve laser action at two lines significantly different in wavelength and gain it is convenient to use an unsymmetric binary cavity, schematically drawn in Fig. 1a. The output mirror M_0 and the grating G form an optical resonator for the wavelength λ_1 which has smaller gain, e.g., a 9 µm CO_2 line. For another wavelength λ_2, e.g., a 10 µm CO_2 line, the grating acts in non-Littrow arrangement and the cavity is closed by the additional mirror M_1. The latter cavity has higher losses and a longer round trip time that compensates higher gain at λ_2. Careful matching of the mentioned parameters allowed simultaneous oscillation at 9 and 10 µm lines in TE CO_2 laser (175 mbar) [17], and even continuous double-wavelength action at low pressure (4 mbar) [18].

1.3 Symmetric Binary Cavity

The configuration of Fig. 1a has some obvious limitations. Turning the grating G will change both λ_1 and λ_2. In most cases, independent wide range frequency tuning, stabilization and loss regulation for every wavelength are desirable. This can be done with non-Littrow arrangement of the grating and its own end mirror for every laser line. Figure 1b shows an example in which all the mirrors have the same radius of curvature and the same distance from the spherical grating G. Such a cavity was already used for a CO_2 laser in [19] and later with some modifications in [20]. In both cases it was used for alternative generation of 9 and 10 µm CO_2 lines. Simultaneous oscillation at two lines in a TEA CO_2 laser with this cavity was achieved in [21]. Because of strong competition, the lines had different transverse mode shapes, i.e., they were spatially separated. The same cavity was also applied for the CO laser [22].

The distance of the additional end mirrors can also be chosen in a way different from Fig. 1b if their radii of curvature are always matched to the wavefronts. In fact, in our experiment we placed two plane mirrors into the mode waists of the branch with separate wavelengths. This is shown in Fig. 2. This figure also indicates the multiply folded beam path in our oscillator as well as the Q-switch, which is a mechanical chopper in the focus of a telescope. For details see [23]. The set-up is based on an industrial 1 kW CO_2 laser

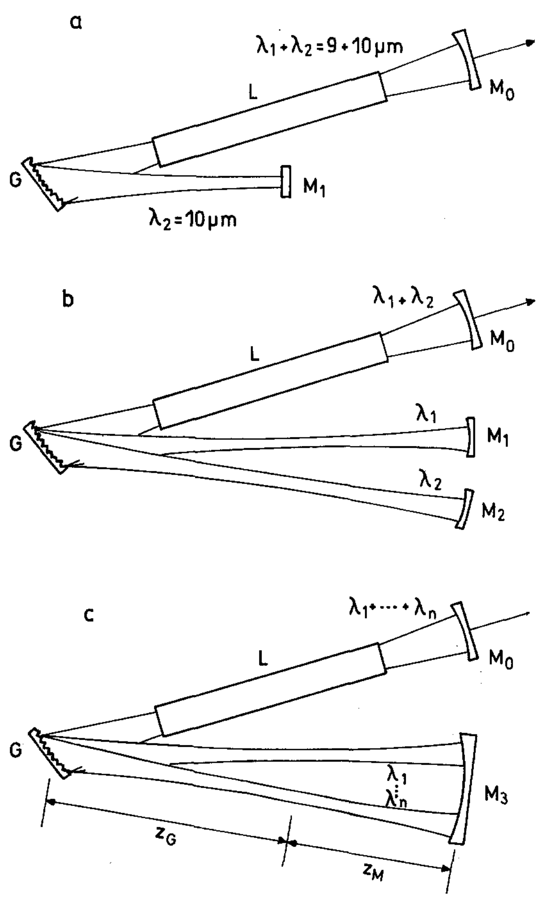

Fig. 1. Multiwavelength resonators: **a** with asymmetric pathway, different losses and roundtrip times for λ_1 and λ_2 (e.g., 9 and 10 µm CO_2 lines); **b** with symmetric pathway for λ_1 and λ_2; **c** with common spherical mirror, supporting multiwavelength oscillation. The laser tube L acts as a diaphragm for transverse mode selection

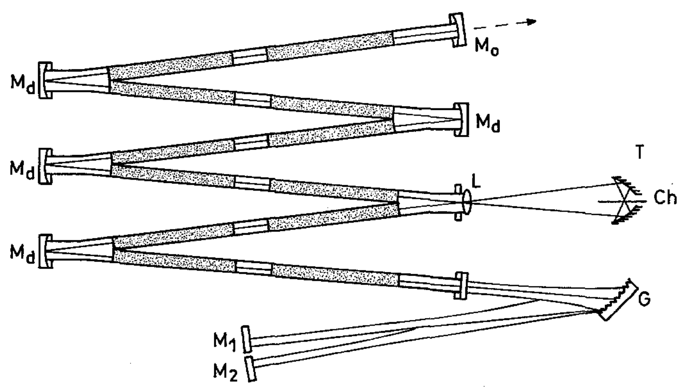

Fig. 2. Setup of the oscillator. The Q-switch is a chopper Ch, rotating in the focus of a parabolic cylindric telescope T. Radius of curvature of the deflection mirrors M_d and of the output mirror M_0: 7.5 m, of the grating G: 6 m, of the tuning mirrors M_1, M_2: ∞, distance M_0–M_d: 3 m, M_d–G: 4.5 m. The waists of the mode are located between the two discharge sections (shaded) of each tube and on M_1, M_2. Rayleigh length $z_0 = 3$ m. Reflectivity of M_0: 3.5%. The grating has 75 grooves per mm

(Ferranti MFK) with slow gas flow, which consists of 12 straight tubes (each containing two discharge sections). We use 6 of these tubes for the oscillator shown in the figure, whereas the 6 others are used as an amplifier (not shown).

Near the end mirrors M_1, M_2 there were two beam stops. By inserting one of them slightly into the beam path, one of the wavelengths can be selectively attenuated in favor of the other. The delay between the pulses of the different wavelengths changes at the same time. This method of controlling the relative pulse energies and delays was introduced by the authors of [14].

1.4 Multiwavelength Cavity with Common End-Mirror

It is suggestive to look for a resonator in which the end mirrors M_1 and M_2 are replaced by a single curved mirror (Fig. 1c). If the end mirror has its center of curvature at the position of the grating, it can obviously support many wavelengths simultaneously. Diaphragms and beam stops in front of it can then serve for selection of the desired laser lines. Its radius of curvature (r_M) must, however, also match the radius r of the wavefront of the desired mode. If the mode is chosen such as in Figs. 1b and 1c, this condition reads

$$r = z_M + z_0^2/z_M = r_M\,, \tag{1}$$

where z_M is the mirror distance from the waist (Fig. 1c) and z_0 is the Rayleigh length corresponding to this waist. The first equality is implied by the general laws of propagation of Gaussian beams [24]. If the grating is at a distance z_G from the waist and if it is in the center of curvature of the mirror, we have

$$z_M + z_G = r_M\,. \tag{2}$$

Equating (1) and (2) yields

$$z_M z_G = z_0^2 \tag{3}$$

as the condition that one common mirror (M_3 in Fig. 1c) supports many different wavelengths simultaneously, all of them having the same transverse mode (the same as for Littrow arrangement).

One can also employ additional focusing elements between G and M_3. For example, one can put a lens of focal length f_L at a distance f_L from the grating. The axes of the beams (which transform like rays in geometrical optics) will then be parallel after the lens. Then for M_3 a plane mirror can be put into the waist of the beams. It will simultaneously return all beams unchanged. Such a resonator, with a concave mirror instead of a lens, has been demonstrated in [25]. It was used for successive tuning from line to line in CO_2 and CO lasers [25, 26]. The design procedure given in [25] is, however, complicated and does not allow to foresee in a simple way the transverse mode dimensions. We recommend, instead, to start out from the shape of the mode in the common branch. This mode is usually desired to fill more or less the active medium. Then after separating the beams by the grating, consider how the Gaussian beams propagate through space and the optical elements (if used). The final mirror should have its center of curvature on the (image of the) grating and at the same time match the curvature of the wavefront.

In the next two sections we describe laser output parameters obtained in practice for one- and two-wavelength oscillation in the symmetric binary cavity and multiwavelength action in the resonator with common end mirror.

2 One- and Two-Wavelength Oscillation

We used the same cavity (Fig. 2) for two- and one-wavelength operation. To generate a single wavelength, one of the end mirrors M_1, M_2 was covered. Closed circles in Fig. 3 show the results of the energy measurements for this case.

Our laser design allows to work with pulse repetition frequency f of more than 20 kHz. The reported data were measured at $f = 8$ kHz, gas pressure of 22 mbar (CO_2 : N_2 : He $\approx$ 20 : 10 : 70) and current of 25 mA per 1 m tube. Under these conditions the laser produces pulses with energy of about 24 mJ at the strongest $9P\,CO_2$ lines. At reduced repetition rate (4 kHz) the energy increases up to 28 mJ. The radiation pulses have a halfwidth of $\tau_{0.5} \approx 200$ ns and a tail of about 0.8 µs containing one fifth of the energy. For weak lines the pulses are longer ($\tau_{0.5}$ up to 300 ns).

The energy observed at the individual laser lines (Fig. 3) was proportional to the energy stored on the respective upper laser states. That is, the tuning curve was similar to the gain spectrum. Such a behavior can be expected for a saturated single-pass amplifier having not too fast rotational relaxation. With the low reflectivity of its output coupler (about 3.5%), our oscillator resembles such an amplifier. This view is also confirmed by the observation that the pulse energy from the oscillator alone was just half the energy of oscillator plus amplifier. (Both of them have the same length of the active medium.)

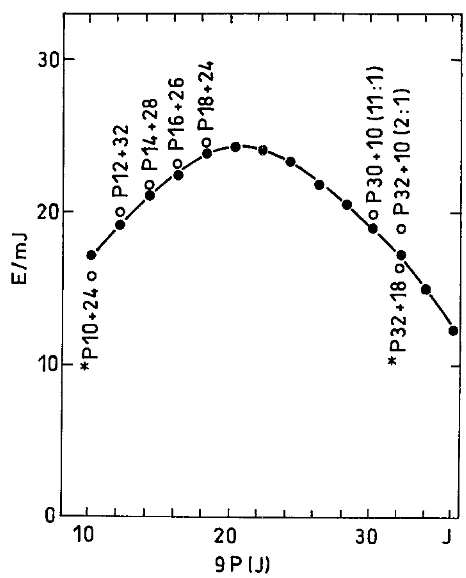

Fig. 3. Tuning curve for one- and two-wavelengths operation. For the latter case, the total energy is shown. The distribution of the energy measured after wavelength separation by a grating monochromator is indicated in the brackets, if it is different from 1 : 1. The resonator design is the same as in Fig. 2

In order to receive a flatter tuning curve, we also tried higher reflectivities of the output coupler (10, 17, and 30%). They should be closer to the optimum for the lower gain lines. However, we did not obtain higher pulse energies on these lines, whereas the emission near $P20$ was weaker. This result points to a nonnegligible loss of the resonator. One loss is certainly the grating: With its efficiency of 90% for the most favorable polarization, it causes 20% loss per roundtrip. Numerous mirrors, windows, matching lens and diffraction phenomena will accumulate to at least another 20% losses.

The open circles in Fig. 3 are the data for two-wavelength pulses. For the pairs of lines with nearly equal gain (symmetric around $9P20$, $9P22$) each wavelength had one half of the total energy with only minor adjustment by the attenuator. In the case of unequal gains we could observe the ratio of energies up to 11:1, e.g., for the $9P30 + P10$ line pair. Choosing lines more asymmetric in gain, two-wavelength oscillation was possible only with attenuation of the stronger line. Such cases are marked by an asterisk in Fig. 3. Because of significant additional losses, the total energy for these line pairs was smaller than for the stronger line alone. In two-line operation without additional attenuation for one of the frequencies, the total pulse energy and peak power was $\sim 10\%$ higher than with a single-line only.

The fact that the difference is small, indicates that the rotational relaxation is fast enough to enable efficient energy extraction on a single wavelength within 200 to 300 ns. Using the published rates [27], the time constant for 22 mbar of our gas mix is $t_{\mathrm{rot}} = 8\,\mathrm{ns}$.

This is the time necessary to refill one rotational level which has suddenly been emptied by stimulated emission. The time to deplete all rotational levels is b^{-1} times longer, where b is the relative population of the emitting state. For the $P20$ line $b = 1/15$ at a temperature of 400 K and the depletion time will be $b^{-1}t_{\mathrm{rot}} = 120\,\mathrm{ns}$.

It is interesting to see that this time is nearly equal to the maximum delay between emission at λ_1 and λ_2, which we observed at extremely different intensities of λ_1 and λ_2. Figure 4 shows the experimental dependence of the delay on the relative intensity of λ_1. Obviously, after about 100 ns the relaxation is so efficient that the first line eats up the inversion of the second line before the latter can grow more.

At first sight it is surprising that in Fig. 4 the time delay is not zero when the pulse intensities are equal and that the delay Δt drops to zero only at strong attenuation of the $P16$ line. To understand such a behavior we consider four competing effects.

1) Laser oscillation sets in when the losses γ have dropped to a level where they are equal to the gain a. This happens already early during the switching time, which for our chopper is about 500 ns. Because of its smaller net gain $(\alpha - \gamma)$, the line $P28$ reaches this threshold later then $P16$. Introducing an additional loss for $P16$ reduces this delay, i.e., *it postpones* $P16$.

2) The buildup-time – that means the time from the begin of the oscillation to the point where the gain starts to drop due to stimulated emission – is inversely proportional to the net gain. This effect in combination with the one described in 1) delays the begin of the laser pulse with lower net gain.

3) The line with the higher net gain has the steeper leading edge. So increasing γ for $P16$ will also slow down its development.

4) If γ for $P16$ is chosen so that the net gains are equal for the two lines, the early parts of the pulses will coincide. But the line with higher gain ($P16$) emits (and loses) photons faster. It will reach the elevated threshold (which coincides with the maximum of the pulse) earlier and attains a smaller peak power. Consequently, even if $P16$ is weaker, its maximum can be reached earlier than the maximum of $P28$.

Furthermore, increased losses decrease the photon lifetime in the resonator and therefore shorten the trailing edge of the laser pulse.

A computer simulation of laser kinetics confirmed this interpretation.

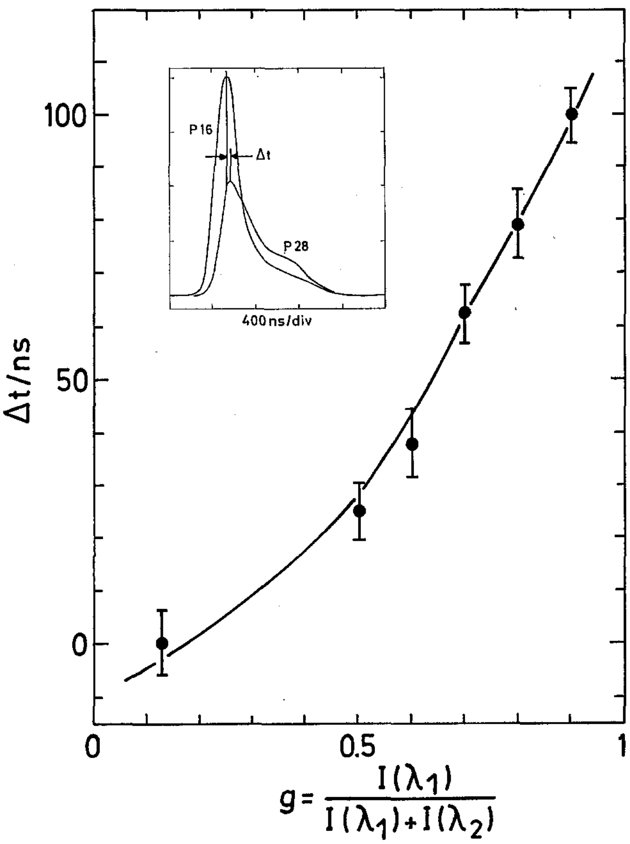

Fig. 4. Time delay, $\Delta t = t(\lambda_2) - t(\lambda_1)$, between the pulse peaks of $\lambda_1 = 9P16$ and $\lambda_2 = 9P28$ vs relative intensity g of the $9P16$ line. The value g was controlled by the attenuators. Without attenuation the delay is about 30 ns at a g value of 0.6. The insert shows typical averaged pulse shapes

3 Multiwavelength Oscillation in the Cavity with Common End-Mirror

To build the desired cavity (Fig. 1c) and satisfy the conditions (2) and (3) we substituted the end mirrors M_1 and M_2 by a spherical mirror M_3 with radius of curvature $r_M = 6\,\mathrm{m}$, at a distance $z_G + z_M = 6\,\mathrm{m}$ from the grating ($z_G = z_M = z_0 = 3\,\mathrm{m}$). The mirror M_3 has a diameter of 10 cm which allows tuning from $9P10$ up to $9P30$. With completely opened mirror M_3 the laser simultaneously oscillates at six lines

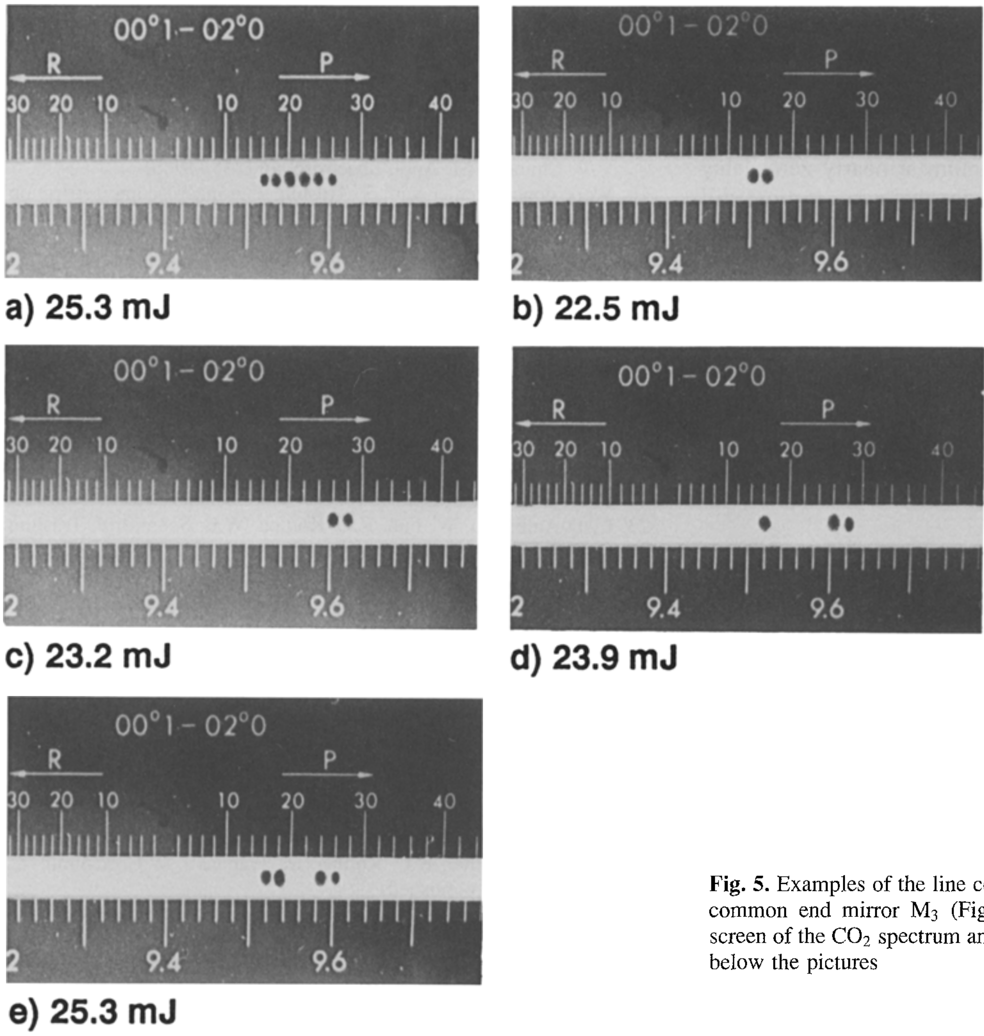

Fig. 5. Examples of the line combinations generated in the cavity with common end mirror M_3 (Fig. 1c). The photos were taken from the screen of the CO_2 spectrum analyser. Total pulse energies are indicated below the pictures

$9P16$–$9P26$ around the strongest $9P22$ line. Figure 5a gives a qualitative picture of this distribution. The total energy of the multiwavelength pulse was 25.3 mJ. This is only slightly more than for two-wavelength operation at $9P18 + 9P24$ lines. There was also no noticeable difference in the pulse lengths. Partial covering of the mirror M_3 initiates oscillation at different line combinations. Adjustment of the several beam stops permits any combination of lines in the tuning range. Some examples and the appropriate pulse energies are shown in Fig. 5b–e.

In our opinion the described cavity is a very good choice if one needs tunable oscillation at three or more close wavelengths, e.g., in the same band and rotational branch of CO_2.

4 Conclusion

Using atmospheric pressure CO_2 lasers, generation of multiwavelength radiation can be obtained either by nonoverlapping beams in one laser module [1] or by combining the radiation of several different units. In the first case, it is difficult to make full use of the laser volume. The second one can suffer from problems with trigger reliability. Furthermore, it is not trivial to combine the two (or more) beams, having them overlap over long path lengths. Our schemes avoid these problems and only one laser is required. The multiwavelength radiation is automatically emitted in one beam.

There are also several limitations of these configurations: The maximum delay is about 100 ns, and it is difficult to obtain oscillation on lines with very different gain. Whereas these features have so far not been a problem for applications, they can in principle also be solved. One can place, for example, the chopper into the waists in the wavelength-separated branch of the resonator, so that the different laser line are switched one after each other. A low gain on one line can be compensated by installing an additional gain tube into the corresponding beam path in the wavelength-separated part.

As a limitation of our scheme one can also consider that the delay between two wavelengths is not independent of their relative power. In laser isotope separation by multiphoton excitation, it is desirable to use a weaker first pulse at a wavelength λ_1, followed by a stronger pulse at λ_2, $\lambda_2 > \lambda_1$. The pulse λ_1 causes isotopically selective preexcitation of molecules, while λ_2 dissociates them. Whereas our scheme – unexpectedly – allow such pulse pairs to a limited extent (Fig. 4), the typical sequence is the other way around.

Furthermore, there is strong temporal overlap in all cases. Multiphoton excitation by two simultaneous wavelengths is fundamentally different to excitation with delayed pulses. As shown in [28], temporal overlap can improve the efficiency of excitation, but degrade the isotopic selectivity. For $CHClF_2$ we observed an optimum at nearly zero delay [29]. So our laser is most suitable for this case. It must still be investigated whether this result can be generalized.

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