

Bistability and Passive Q-Switching of a CO, Laser with Saturable Absorber

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Abstract. The coexistence of bistability and repetitive passive Q-switching of a $CO₂$ laser with a gaseous saturable absorber has been investigated experimentally. With $CH₃F$ as the saturable absorber bistability has been observed simultaneously with passive Q-switching, whereas with $CH₃OH$ either one of these phenomena can only exist, as demonstrated for the first time. The measured features of both systems can be explained qualitatively by rateequation theory.

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Lasers with saturable absorbers are known to show repetitive passive Q-switching (PQS) and bistability, depending on operating conditions. These phenomena have been studied separately quite extensively in the past, especially for gas lasers with saturable absorbers $[1–6]$. Passively Q-switched molecular gas lasers have found a number of applications, e.g. for spectroscopic purposes in intracavity molecular absorption studies [7] or for the pumping of FIR lasers, in both extracavity [8] and intracavity configurations [9].

Recently, there have been several theoretical [10-12] and experimental attempts $\lceil 12 - 14 \rceil$ to study both PQS and bistability simultaneously in a laser with saturable absorber. The reason for these investigations is the large variety of phase-transition-like phenomena in the laser with saturable absorber which may be of interest from the view point of synergetics [15] and for the study of dissipative systems [16]. Up to now the theory of the laser with saturable absorber seems to be more developed than the respective experiments. It was therefore the aim of the present investigation to provide some more quantitative experimental data on simultaneous PQS and bistability for a comparison with existing theory.

1. Theoretical Considerations

In lasers exhibiting PQS the field strength is changing slowly compared to the cavity round-trip time. Therefore, population effects are considered to be dominant in this system and a rate-equation approximation should be adequate. Such a theory, being applicable for the $CO₂$ laser with a molecular gaseous absorber has been published by Arimondo et al. [12]. It will be shown that this theory is sufficient for a qualitative interpretation of the observed features. Both, the amplifier and the absorber are treated in this theory as a 4-level system, which is adequate for IR absorbing molecules, where the optical transition occurs between two rotational levels in different vibrational states. The rotational levels are coupled by dipole collisions to the rest of the rotational levels in the same vibrational state. The vibrational relaxation occurs via binary collisions or by diffusion. Usually the vibrational relaxation is about three orders of magnitude slower than the rotational relaxation, leading to the wellknown "vibrational bottleneck" effect [17] in optically pumped FIR lasers. It has been shown by Arimondo et al. that the 4-level model gives a more realistic description of PQS in molecular gases than a 2-level model. Effects which are not included in the 4-level model are the detuning of the laser field from the absorption resonance and the inhomogeneous broadening of the optical transitions.

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The most important results of the 4-level theory are summarized as follows:

In order to obtain the bistability condition only the stationary solution of the rate-equation system has to be considered, leading to the usual laser equation

$$
2KI_{st}\left(1-\frac{A}{1+I_{st}}+\frac{\bar{A}}{1+aI_{st}}\right)=0,
$$
\n(1)

where I_{st} is the normalized stationary laser intensity, $2K$ the cavity loss, A the small-signal gain divided by $2K$, \overline{A} the corresponding absorber parameter, and \overline{a} is the ratio of the saturation intensities of the amplifier and the absorber. For $A \leq (A+1)$ this equation has a positive nonzero solution I_+ if

$$
a>1
$$
 and $\bar{A}>1/(a-1)$. (2)

In this case the laser is bistable between $I_{st} = 0$ and $I_{st} = I_{+}$. Varying a control parameter, e.g. the gain, the laser intensity goes through a hysteresis loop with an abrupt first-order-like phase transition, switching from one solution to the other.

From linear stability analysis, the stability of the rateequation solution can be deduced, leading to the condition for PQS. Under the condition that $K \gg \gamma$, $\bar{\gamma}$, where γ , $\bar{\gamma}$ are the vibrational relaxation rates of the amplifier and the absorber, respectively, two different instability conditions are derived. For $I_+ < 1$ the instability condition is

$$
A \lt \xi a \overline{A},\tag{3a}
$$

where ξ depends on the relaxation rates of the amplifier and the absorber.

For $I_{+} > 1$, which means strong saturation of the amplifier and the absorber, the condition is simply

$$
aA < \overline{A} \tag{3b}
$$

It should be noted that inequality $(3a)$ has usually been used only to describe PQS. From (3a) and (2) simultaneous PQS and bistability can be predicted easily, whereas in the case of strong saturation the two phenomena are mutually exclusive due to (2) and (3b).

The experiments reported here, like previous experiments, were carried out with a saturable absorber at low gas pressure, where Doppler broadening is dominant. Thus only qualitative agreement between theory and experiment can be expected.

2. Experiment

The experimental set-up has been described in detail in [18]. It consists of a FIR-CO₂ hybrid laser which was operated for the present investigation as a $CO₂$ laser

only, the FIR laser part being used as an intracavity absorber cell. The amplifier is a conventional $CO₂$ laser tube (Coherent model 42) with two separate discharges of 40 cm length each. The absorption path length is 1.4 m and the overall cavity length is 2.6 m. The $CO₂$ laser radiation is coupled out through a Ge mirror with 2% transmittance. The laser output was detected by a power meter and a fast, room-temperature HgCdTe detector. The parameters varied in the experiment were the pressure of the $CO₂$ laser gas mixture (12:18:70 parts of $CO_2:N_2$:He), the $CO₂$ laser discharge current and the absorber gas pressure. The cavity losses could be varied too by closing an intracavity iris diaphragm. The measurements were performed with the $CO₂$ laser operating in the free running mode. For this purpose the laser cavity length was set by a PZT for maximum output power, a point which was consistent also with most stable PQS pulses. To avoid drifts in cavity length, warm-up times of the system of several hours have been used.

3. Results

3.1. CH₃OH

We have found bistability and PQS with methanol as the absorber for the $CO₂$ laser operating on the 9P(16), 9P(34) and 9P(36) lines. Since the results for all three lines are quite similar, only the measurements on 9P(36) are reported here.

Figure 1 shows the output power of the laser as a function of discharge current for different absorber pressures. At zero absorber pressure the laser is monostable and operates cw only. At $4 \text{ Pa } CH_3OH$, the laser is monostable and passively Q-switched at low currents but bistable and cw lasing at high currents. At an absorber pressure of 7 Pa the laser operates monostable in the PQS mode over the whole current range. It is worth noting that the threshold for the laser oscillation in Fig. 1 depends more on the absorber pressure at the high current than at the

Fig. 1a-c. $CO₂$ laser power vs. discharge current for different $CH₃OH$ pressures: 0Pa (a), 4Pa (b), and 7Pa (c)

low-current side. This indicates that the small-signal gain is decreasing with current while the saturation intensity is increasing. This point will be important in the following discussion.

The main feature of $CH₃OH$ is observed more clearly from Fig. 2, which shows the laser power versus absorber pressure for different discharge currents. At a laser current of 50 mA the laser operates cw at low pressure. At about 3.2 Pa the laser starts with passive Q-switching and the output power decreases slowly with pressure. At higher current (60 mA) this decrease of power in the PQS range is steeper and at 70 mA the laser goes directly from the cw to the off-state. Together with this abrupt change in output power there appears a hysteresis on the pressure scan, demonstrat-

Fig. 2a-e. $CO₂$ laser power vs. CH₃OH pressure for different discharge currents: 50 mA (a), 60 mA (b), 70 mA (c), 80 mA (d), and 90 mA (e)

ing the bistability in this case. The hysteresis cycle becomes even larger at higher currents (80-90 mA).

Figure 2 shows clearly that either PQS or bistability can take place for $CH₃OH$ as the absorber. The parameter ranges for the different modes of laser operation at different $CO₂$ laser gas pressures and with additional cavity losses introduced by the iris diaphragm are depicted in the phase diagrams Fig. 3a–e. As it is usually expected, PQS is found with increasing absorber pressure. Bistability occurs here at high laser currents between the cw and the off-state only. It is worth noting that the mutual exclusion of PQS and bistability, which is reported here for the first time, is relatively insensitive to the cacity loss, e.g. by closing the iris diaphragm the laser cavity loss has been increased from about 15% to 30%. A more detailed discussion of the results can be given only if the experimental parameters of the amplifier and the absorber are known. The determination of these parameters and the discussion are given in the Sects. 4 and 5.

3.2. CH₃F

Operating the $CO₂$ laser on the 9P(20) line with methyl fluoride as the absorber, a bistability region is again observed, as shown in Fig. 4. In contrast to the experiments with $CH₃OH$, however, the bistability occurs between the PQS and the off-state. It should be mentioned that the hysteresis cycle observed here is in complete qualitative agreement with the theory showing abrupt switching at the end points of the cycle in contrast to findings in a previous experiment [12]. The parameter ranges for the different states of laser

Fig. 3a-e. Phase diagrams of the CO₂ laser with CH₃OH as saturable absorber for different CO₂ laser gas pressures: 1.3 kPa (a), 2.6 kP a (b), 3.3 kP a (c). Introducing an intracavity iris diaphragm, phase diagrams are recorded for the pressures: 2.6 kP a (d) and 1.3 kP a (e)

operation with $CH₃F$ as the absorber are depicted in the phase diagrams shown in Fig. 5a-c. While PQS is observed at high pressures, as expected, bistability is found again at high currents, but in contrast to the experiments with CH₃OH now at relatively high absorber pressures only. The reason for this behaviour can only be discussed if the amplifier and the absorber parameters are at least qualitatively known.

4. Determination of the Experimental Parameters

The parameters needed for a qualitative comparison with the theoretical model are the small signal gain and the absorption coefficients of the amplifier and the absorber, respectively, as well as their saturation intensities. If the absorber parameters are known, the

Fig. 4a and b. $CO₂$ laser power vs. discharge current for different $CH₃F$ pressures: 0Pa (a) and 33Pa (b)

gain parameters can be calculated from the phase diagrams and the output intensity of the $CO₂$ laser at zero absorber pressure. We therefore measured the absorption in $CH₃OH$ and $CH₃F$ and found small signal absorption coefficients at the $CO₂$ laser line centers of $0.011 \pm 10\%$ (m \cdot Pa)⁻¹ for CH₃OH at the 9P(36) line and $0.010 \pm 10\%$ (m \cdot Pa)⁻¹ for CH₃F at the 9P(20) line. While the value for $CH₃OH$ is found to be somewhat higher than in [19], the value for $\rm CH_3F$ is somewhat lower than in [19, 20]. The saturation intensity for $CH₃OH$ was measured at low pressure, giving a value of $0.02 + 20\%$ W/(cm \cdot Pa)². No saturation could be measured at low pressures in CH_3F with intensities of $15 \,\mathrm{W/cm^2}$, thus in agreement with [19, 20] which gives values for the saturation intensity at low pressure in $CH₃F$ between 0.13 and $0.45 W/(cm \cdot Pa)^2$. For pressures above 7 Pa a deviation from the quadratic pressure dependence of the saturation intensity in $CH₃F$ has been reported [19]. The saturation intensity only slightly increases with pressure up to about 70 Pa due to the slow vibrational relaxation in $CH₃F$ [21].

Using the measured small-signal absorption coefficients, the small-signal gain of the amplifier can be calculated from the laser threshold in the phase diagrams. The small-signal gain coefficients for the $9P(36)$ and the $9P(20)$ lines as a function of discharge current are shown in Fig. 6a and b. In both cases the gain shows a maximum at relatively low currents and falls off almost linearly at higher currents. From Fig. 6 the cavity loss $2K$ has been estimated to be about 15%, which is a realistic value since the laser grating, in our case, is used as a folding mirror in the cavity. The

Fig. 5a-c. Phase diagrams for the CO_2 laser with CH₃F as saturable absorber for different CO_2 laser gas pressures: 1.3 kPa (a), 2.6 kPa (b), and 3.3 kPa (c)

Fig. 6a and b. Small-signal gain coefficients vs. discharge current for different CO_2 laser gas pressures with the laser operating on the $9P(36)$ line (a) and the $9P(20)$ line (b)

Fig. 7a and b. Saturation intensity vs. dicharge current for different CO₂ laser gas pressures with the laser operating on the 9P(36) line (a) and the 9P(20) line (b)

saturation intensity of the $CO₂$ laser then has been calculated from the output power of the laser, assuming homogeneous broadening at all laser gas pressures. The results are shown in Fig. 7a, b. In contrast to the small-signal gain, the saturation intensity is increasing monotonically with current in agreement with earlier work on CO_2 lasers [22]. With the dependences of the amplifier and the absorber parameters on discharge current and absorber pressure given here, the experimental results now can be discussed qualitatively as follows.

5. Discussion of Results

5.1. **CH₃OH**

The bistability in methanol is found at high laser currents and low absorber pressures. This can be understood from the bistability condition (2). The relative saturability a is increasing with current but

proportional to $1/p^2$, p being the absorber pressure. Since \overline{A} is increasing only linearly with p it is clear that, if bistability occurs at all with $CH₃OH$ as the absorber, it will occur at high currents and low pressures. PQS in $CH₃OH$ can be described either by condition (3a) or (3b) depending on the laser intensity. For the mutual ecxlusion of PQS and bistability, the state of the laser is certainly described by the situation at the transition from cw to pulsed emission. It can be seen in the phase diagrams that this transition can only be described by (3b) since the transition point from cw to PQS is going to higher absorber pressures in a region of decreasing A and increasing laser saturation intensity. Assuming therefore the high intensity condition (3b) to be the relevant condition for PQS in $CH₃OH$, the mutual exclusion can be easily explained. While PQS is expected for a $\lt 1$, bistability only can occur for $a > 1$. Beyond that, PQS is expected to be more probable at low current and high absorber pressure from condition (3b), while the reverse is expected for the bistability range as stated above.

5.2. CH₃F

The situation in methyl fuoride is somewhat different from that in methanol. The relevant question here is not whether PQS is determined by condition (3a or b) but rather why the bistability is found at high absorber pressures. This experimental finding only can be explained by assuming existence of a vibrational bottleneck in $CH₃F$ which drastically reduces the absorber saturation intensity compared to the usual quadratic pressure dependence. Such a behaviour has been measured before by Weiss [19]. We therefore believe that the bistability between PQS and the off-state is another manifestation of the vibrational bottleneck-effect in $CH₃F$. While PQS is mainly determined by the fast rotational relaxation, the bistability is determined by the decrease in the vibrational population difference due to the slow vibrational relaxation.

6. Conclusions

The coexistence of passive Q-switching (PQS) and bistability has been investigated experimentally in a $CO₂$ laser with CH₃OH or CH₃F as the saturable absorber.

While in the case of $CH₃OH$, mutual exclusion of PQS and bistability has been observed for the first time, simultaneous PQS and bistability has been observed with $CH₃F$ as the saturable absorber. It has been shown that the mutual exclusion of PQS and bistability in $CH₃OH$ can be explained if strong saturation of both, the amplifier and the absorber, is assumed for the condition of $POS - an approximation to describe$ PQS, which never has been used before. The finding in $CH₃F$ reflects the influence of the vibrational relaxation on the absorption in $CH₃F$, a fact which is important for optically pumped FIR lasers. Although a qualitative agreement between theory and experiment could be established using a theory for homogeneously broadened transitions it certainly would be worth to generalize the theory to inhomogeneous broadening. In this case a quantitative description of PQS and bistability in gas lasers with gaseous saturable absorbers at low pressures should be possible.

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References

- 1. I. Burak, P.L. Houston, D.G. Sutton, J.I. Steinfeld: IEEE J. QE-7, 73-82 (1971)
- 2. H.T. Powell, G.J. Wolga: IEEE J. QE-7, 213-219 (1971)
- 3. J.Dupr6, F. Meyer, C. Meyer: Rev. Phys. Appl. (Paris) 10, 285-293 (1975)
- 4. A.P. Kazantsev, S.G. Rautian, G.I. Surdutovich: Sov. Phys. JETP 27, 756-762 (1968)
- 5. L.A. Lugiato, P. Mandel, S.T. Dembinski, A. Kossakowski: Phys. Rev. A 18, 238-254 (1978)
- 6. S. Ruschin, S.H. Bauer: Chem. Phys. Lett. 66,100-103 (1979); Appl. Phys. 24, 45-48 (1981)
- 7. W.A. Kreiner: IEEE J. QE-12, 16-20 (1976)
- 8. J.W. Won, G.D. Willenberg: Appl. Phys. B31, 1-4 (1983)
- 9. G.A. Koepf: Appl. Phys. Lett. 31,272-273 (1977)
- 10. T. Erneux, P. Mandel: Z. Phys. B 44, 353-374 (1981)
- 11. J.C. Antoranz, J. Gea, M. Velarde: Phys. Rev. Lett. 49, 35-38 (1982)
- 12. E. Arimondo, F. Casagrande, L. Lugiato, P. Glorieux: Appl. Phys. B 30, 57-77 (1983)
- 13. A. Jacques, P. Glorieux: Opt. Commun. 40, 455-460 (1982)
- 14. J.W. Won: Opt. Lett. 8, 79-81 (1983)
- 15. H. Haken: *Synergetics- An Introduction,* 2nd ed. (Springer, Berlin, Heidelberg, New York 1977)
- 16. G. Nicolis, I. Prigogine: *Self-Organization in Non-Equilibrum Systems* (Wiley, New York 1977)
- 17. D.T. Hodges, J.R. Tucker, T.S. Hartwick: Infr. Phys. 16, 175-182 (1976)
- 18. Z. Solaji6, J. Heppner: Appl. Phys. B 33, 23-27 (1984)
- 19. C.O. Weiss: IEEE J. QE-12, 580-584 (1976)
- 20. D.T. Hodges, J.R. Tucker: Appl. Phys. Lett. 27, *667-689* (1975)
- 21. E. Weitz, G. Flynn, A.M, Ronn: J. Chem. Phys. *65, 6060-6067* (1972)
- 22. J. Tulip: IEEE J. QE-6, 206-211 (1970)