Appl. Phys. B 64, 423–427 (1997)

Applied Physics B Lasers and Optics

Study of simultaneous oscillation on several transitions in cw CO laser

V.A. Gorobets, V.O. Petukhov, S.Ya. Tochitsky, V.V. Churakov

Institute of Physics, Academy of Sciences of Belarus, Minsk 220072, Belarus

Received: 13 September 1995/Revised version: 5 August 1996

Abstract. The stability of simultaneous oscillation on several lines in a cw CO laser with a multichannel cavity was studied for different combinations of rotational-vibrational lines. The field mechanism approach was applied for explanation of the peculiarities of laser oscillation. The criteria for selection of the optimal lines for stable multiline oscillation were determined. Based on these investigations a stabilized three-color CO laser was developed and utilized for nondestructive testing of the quality of industrial silicon ingots.

PACS: 42.60; 42.55E; 07.65

Laser sources having simultaneously two or more wavelengths in the output are required in various fields. For example, lasers oscillating on several rotational-vibrational transitions in the mid-IR are of interest for vibrational excitation and dissociation of molecules, remote and local gas analysis, metrology, nonlinear conversion in crystals, and effective optical pumping of FIR lasers [1, 2]. However, in the case of molecular gas lasers including cw CO lasers, achieving simultaneous oscillation on several given lines, with stable output parameters, is a complicated problem, mostly because of the strong competition between rotational-vibrational transitions.

It is known that a cw CO laser with a nonselective cavity oscillates spontaneously on several (up to 40) rotational-vibrational transitions [3]. The spectral distribution of the output is changed at random due to competition between these transitions. Two main processes are at the heart of this competition effect: first, a deviation from the Boltzmann rotational distribution or quasi-equilibrium distribution of particles among vibrational levels by the laser field; second, rotational or vibrational relaxations that attempt to restore the equilibrium state. To obtain stable cw oscillation on several given lines simultaneously, it is necessary to choose the CO laser parameters correctly, considering the competition and relaxation processes.

It is natural to assume that simultaneous oscillation on several rotational-vibrational transitions in a CO laser is similar to multiline oscillation in CO_2 laser, which has been studied in much more detail. In our recent papers, on simultaneous oscillation of two and more rotational-vibrational lines in a pulsed TEA CO_2 laser, it was shown both theoretically and experimentally that stable multiline operation is possible at high laser intensities. Under these conditions, a rotational bottleneck effect means the competition influence on the laser stability is weakened considerably [4–8]. To moderate the competition it is necessary for the intracavity intensity to reach the saturation intensity, I_S, for the transition. This intensity can be estimated from the expression

$$I_S = c\pi\gamma/\tau B, \qquad (1)$$

where c is the speed of light, γ is the collisional half-width at half-maximum, τ is the collisional relaxation time, and B is the line center Einstein coefficient for the rotational-vibrational transition. This mechanism, which we call the "field mechanism", is applicable to rotational transition of both one band and various bands, including non-regular bands 00^02-02^01, 00^02-10^01, 01^11-11^10. In the last case it is simplest to moderate the competition influence on laser stability. In this case the vibrational bottleneck effect operates at significantly smaller values of the saturation intensity because of slower vibrational relaxation processes.

Although we studied multiline oscillation in a TEA CO_2 laser, the temperature model used in [8] for calculations is also applicable to cw CO_2 lasers. On the other hand our results are in good agreement with earlier theoretical works on multiline oscillation in a cw CO_2 laser [9, 10]. Calculations [9, 10] have shown that in order to weaken the influence of competition among transitions with Lorentzian-shaped line broadening (homogeneous broadening), it is necessary to generate conditions such that the laser field disturbs the Boltzmann rotational distribution. Thus, in our opinion, the field mechanism is universally applicable and can be used to explain the multiline oscillation in both TEA CO_2 (pulse duration of approximately 1 μs) and other cw molecular gas lasers with Lorentzian line broadening. It should be noted that for lasers with Doppler line broadening, different volumes of the active medium can be responsible for the oscillation on different lines.

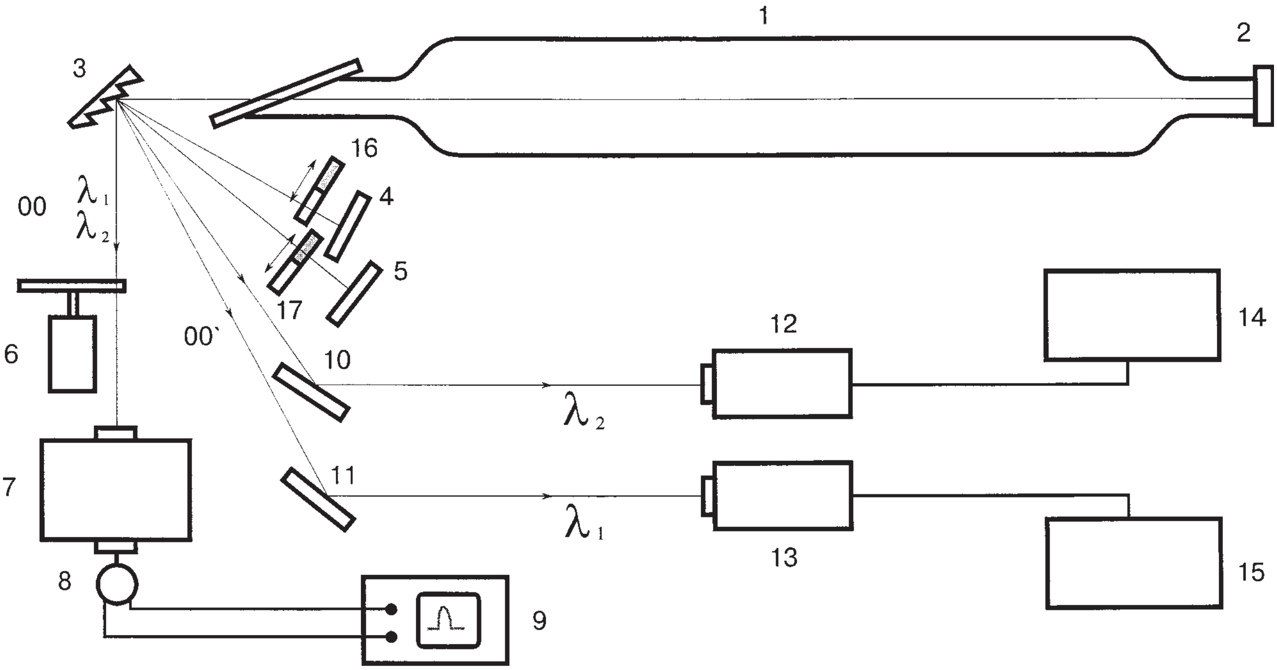

Fig. 1. Schematic diagram of the experimental set up: 1 – discharge tube; 2 – rear mirror; 3 – diffraction grating; 4,5 – additional cavity mirrors; 6 – chopper; 7 – monochromator; 8 – pyroelectric detector; 9 – oscilloscope; 10,11 – steering mirrors; 12,13 – calorimeters; 14,15 – chart recorders; 16,17 – screens*

In this work the field mechanism approach is applied to explain the simultaneous oscillation on several lines in a cw CO laser where the line broadening is close to homogeneous. The stability of this system has been studied in a multichannel cavity with the mutual influence of lasing channel taken into account. To explain the peculiarities of the multiline oscillation in the CO laser, the competition effect has been studied for various combinations of rotational-vibrational lines over a wide spectral range. To demonstrate the principle of multiline laser systems, we present a stabilized three-color CO laser, designed according to the conclusions of this investigation.

1 Experimental set up

The experimental set up for investigating the simultaneous oscillation on two lines is schematically shown in Fig. 1. The experiments were conducted using a commercial sealed-off discharge tube, GL-509, cooled by water. The length of the active medium was ≈ 1.2 m. The discharge tube was powered by a stabilized high-voltage power supply. The cavity was formed by a totally reflected mirror, 2, installed in the tube, a 200 lines/mm grating, 3, operating in first order in a non-Littrow arrangement, and mirrors, 4 (λ_1) and 5 (λ_2) set at small angles to the cavity axis. The reflectivity in the first diffraction order was greater than 94%. The laser output was approximately equally distributed between the 00 and 00′ orders of the diffraction grating (see Fig. 1). When this system operates with the 00 order of the grating, radiation of different wavelengths, λ_1 and λ_2, propagate along the same optical path. Radiation output using the 00′ order provides spatial separation between the λ_1 and λ_2 components [11] so that we could investigate multiline oscillation. Using this arrangement for the cavity it was possible to tune independently the lasing channels to a desired line and analyze the oscillations on several lines simultaneously; in our case, up to four lines when additional mirrors were used.

With the help of additional mirrors, 10 and 11, the 00′-order output was directed to IMO-2N laser calorimeters, 12 and 13, whose signals were observed with the pair of LKS-4-003 roll chart recorders, 14 and 15. For short-term stability measurements (in the microsecond range) we used a Ge:Au detector (cooled by liquid nitrogen) with a bandwidth of ≈ 10 MHz instead of the calorimeter. The electrical signal from the detector was recorded by a digital, two-channel, oscilloscope, 9. To analyze the spectral characteristics of the laser, the 00-order radiation modulated by an electromechanical chopper, 6, and directed through a SPM-2 monochromator, 7, was utilized.

2 Results and discussion

An investigation into the stability of simultaneous oscillation on various transitions was carried out for various pairs of lines belonging to different vibrational bands from $v = 8 \rightarrow v = 7$ to $v = 22 \rightarrow v = 21$ ($\lambda = 5.3$–$6.4\,\mu$m). The vibrational (v) and the rotational (J) quantum numbers were varied. Typical results from the long-term stability measurements over several minutes are given in Figs. 2 and 3. To investigate the effect one lasing channel has on the other the following procedure was used. Two channels were adjusted to the lines at λ_1 and λ_2 by alignment of mirrors 4 and 5. Then the intracavity plates 16 and 17, for radiation screening, were placed into the cavity in front of the mirrors 4 and 5, the chart recorders were

*The 00′ light originates from the radiation which comes from the cavity mirrors 4 and 5. When this radiation hits the grating the major fraction is rediffracted into the laser tube but a small portion is regularly reflected and can be used for diagnostic purposes.

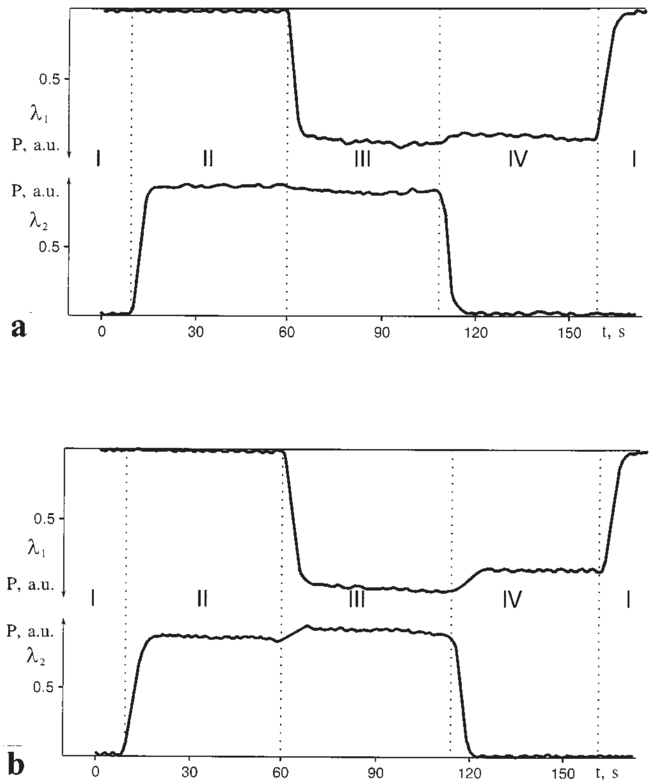

Fig. 2a,b. Recordings of variations in double-wavelength output for lines belonging to different vibrational bands: **a** noncascade transitions ($\lambda_1 = 5.67\,\mu\text{m}$ (13-P(18)), $\lambda_2 = 6.01\,\mu\text{m}$ (17-P(18))); **b** cascade transitions ($\lambda_1 = 5.67\,\mu\text{m}$ (13-P(18)), $\lambda_2 = 5.75\,\mu\text{m}$ (14-P(18)))

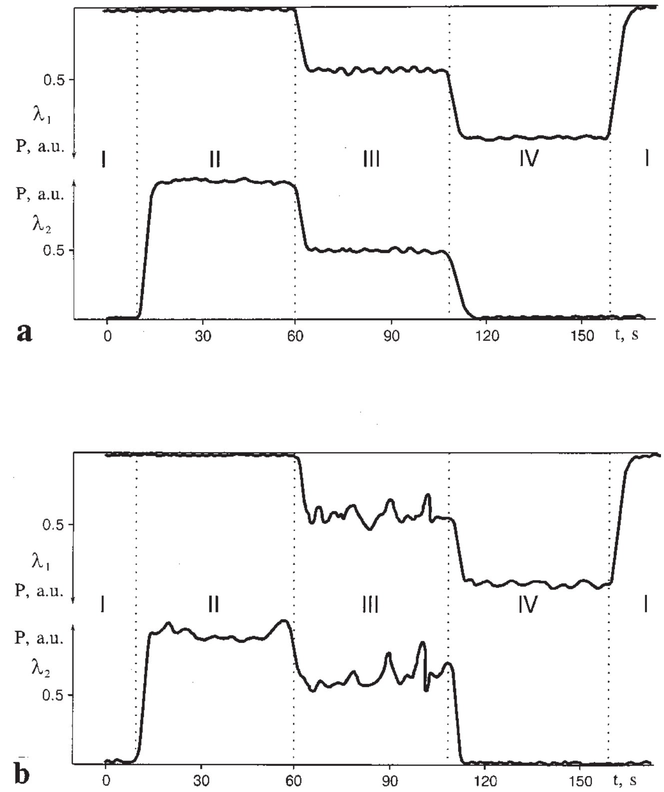

Fig. 3a,b. Recordings of variations in double-wavelength output for lines belonging to a common vibrational band ($\lambda_1 = 5.63\,\mu\text{m}$ (13-P(15)), $\lambda_2 = 5.7\,\mu\text{m}$ (13-P(20))): **a** optimal current; **b** nonoptimal current

then switched on. This time interval is characterized by the absence of oscillation in both channels (zero level) and is labeled I in Figs. 2 and 3. Next the screen blocking the λ_2 channel was quickly removed and the corresponding calorimeter began to record the output power in this channel. After $\approx$ 1 min from the beginning of measurements the second lasing channel, with λ_1, was also opened. Then, closing the lasing channels in turn, we recorded the laser output, starting with the λ_1 channel. According to this procedure the time period labeled II corresponds to channel λ_2 open, III corresponds to both channels open, IV corresponds to channel λ_1 open.

Studying the interaction between the lasing channels with both types of meters (IMO-2N and Ge:Au) has shown that tuning the cavity mirrors to various vibrational bands, independently of the rotational linen choice, leads to stable multiline oscillation. For transitions that are not coupled through a common vibrational state, the output power, as illustrated in Fig. 2a, is essentially independent of the presence of oscillation in the neighboring channel. In the case of cascade transitions the mutual influence of the channels becomes more noticeable (Fig. 2b). For these transitions the output power of each channel increases by $\approx 10\%$ compared with the single-line operation. The increase that takes place in the case of cascade oscillation can be attributed to the population inversion increasing in the adjacent vibrational bands. More detailed investigations of multichannel cascade oscillation may be used to study the vibrational exchange constants and population distribution over vibrational levels in the CO molecule excited state. This is yet important for the development of powerful laser systems [3, 12].

The most significant interference between the channels was observed with lasing from various rotational lines of the same band (Fig. 3). In this case the competition resulted in a decrease in the output power (by nearly one half) in each channel, as opposed to the case of cascade oscillation where the output power increased. At the same time the stability of the oscillation worsened greatly. This was particularly noticeable in the short-term stability measurements from the Ge:Au photodetectors on a microsecond time scale. Even a small misalignment of one of the cavity mirrors or a non-optimum setting of the discharge current caused strong antiphase pulsations in the lasing channels. These pulsations were also observed with the slower IMO-2N calorimeter (see Fig. 3b).

The observed difference in the effect of competition on the interference between the channels for various combinations of rotational-vibrational lines can be explained on the basis of the field mechanism criteria introduced in (1). To do this, one compares the value of the saturation intensity I_S, which depends on the transition probabilities and relaxation rates, with the laser field intensity achieved in the cavity. The high probability of spontaneous emission ($200–300\,\text{s}^{-1}$) and the comparatively low speed of the v-v collisional exchange for the CO molecule ($\approx 10^5\,\text{s}^{-1}\text{Torr}^{-1}$) means, that even the comparatively low intracavity intensity I achieved experimentally (according to our estimates, $I = 20–40\,\text{W/cm}^2$) is much in excess of the saturation intensity I_S defined by (1). As a result the vibrational bottleneck effect comes into action , and the channels of oscillation on the different vibrational transitions can be thought of as relatively independent. The rate of rotational relaxation is several orders of magnitude higher than the rate of vibrational relaxation (about of 10^3 times higher for experimental conditions), and the radiation intensity I is inadequate to produce the rotational bottleneck ($I \ll I_S$) and consequently for reducing the competition between rotational transitions.

A theoretical analysis is required to explain the observed dependencies more accurately. However, the detailed numerical analysis of multiline oscillation in the CO laser, as was done for the TEA CO_2 laser in [8], is complex owing to the quasi-equilibrium vibrational distribution in the electrical discharge. Also, until very recently, there was an absence of accurate data on the relaxation rate for CO until [3, 12]. Estimates of the output-power stability of the multiline CO laser carried out on the basis of the four-level model, developed for the cw CO_2 laser in [9], confirmed that the field-mechanism approach is valid in the situations considered.

Experimental investigations have shown that similar behavior in a multiline cw CO laser also occurs when it is simultaneously oscillating on more than two lines.

3 Application of multiline cw lasers

Understanding the physical processes involved in simultaneous oscillation on more than one line allows one to choose the correct active medium, pumping and resonator parameters to design an effective multicolour laser system with stable output characteristics. As an example of the application of a multiline cw CO laser, we now present a description of a three-colour CO laser spectrometer, developed for measuring impurities in industrial ingots and plates of silicon. The existing electrical and common spectroscopy methods of nondestructive testing do not have the accuracy and spatial resolution currently required, and generally they can measure only one type of impurity. When one probes silicon with the two wavelengths with maximum separation in the CO laser output $\lambda \approx 5\,\mu m$ and $\lambda > 6\,\mu m$), it is possible to determine concentrations of doping impurities with high accuracy, by using the dependence of their absorption on the wavelength [13]. Additional probing at wavelengths near 5.8 µm enables one to measure the concentration of oxygen, which has a strong absorption band in this spectral range [14].

The sealed-off CO discharge tube described above was used as the active element. A cavity scheme similar to that in Fig. 1 was used. An additional mirror to create the third lasing channel was added. To provide passive stabilization, all components of the cavity and the discharge tube were rigidly fixed on three invar rods, as in [15]. The cavity was tuned to three CO rotational-vibrational lines: $\lambda_1 = 5.389\,\mu m$ (9-8 P(20)), $\lambda_2 = 6.178\,\mu m$ (19-18 P(17)), and $\lambda_3 = 5.809\,\mu m$ (15-14 P(16)). These lines were chosen to give high stability in the laser output.

The diffraction grating was used in the 00 order and the three waves propagated along the same direction, which is important for a good spatial resolution of the parameters under study. The total output power was approximately 600 mW and was almost equally distributed between the three wavelengths. The diameter of the output beam was 5 mm ($1/e^2$ level for the TEM_{00} mode in each channel) and had a Gaussian shape. The laser beam was compressed by a factor of 3 with the help of a telescope; this increased the fraction of radiation passing through the adjustable slit in front of the silicon ingot.

An important problem in these measurements is separation of the signal wavelengths after the beam has passed through the sample. To simplify the data gathering and processing it is reasonable to use temporal separation of the signal instead of spatial separation, as this results in improvement of the measurement accuracy. We realized and studied two methods to separate the signals. Figure 4 illustrates one of them. A series of pulses belonging to different waves were produced when an external rotatable diffraction grating was mounted so as to direct the three-color beam onto a photodetector. The best results for signal processing were obtained with a 150 lines/mm grating (blaze angle 45°) operated in first order.

The second method is basted on the use of a chopper made of a light-weight material placed in the laser cavity in front of the additional mirrors. The chopper has holes placed at different distances for each wavelength. The distances from the axis of rotation and the hole diameters were calculated so that as the disc rotates, pulses of the three lines are generated separately. This method was simpler and easier to use than the one mentioned above, but the stability of the laser output was slightly reduced. For example, when the frequency of the chopper rotation was 50–60 Hz the scatter of the pulse amplitudes reached 5%, whereas in the first method this parameter generally didn't exceed 3%. This may be attributed to the pulse periodic mode of oscillation (quasi-continuous wave) in this case.

On the basis of these investigations, several three-color CO laser spectrometers have been produced. They were successfully utilized for rapid measurements of the concentrations of doped impurities and oxygen in untreated industrial silicon ingots with a high spatial resolution (up to 50 µm). The results of these semiconductor measurements will be published elsewhere [16].

One other possible application of multiline systems is using them to study nonlinear dynamics processes. Singleline cw CO_2 and CO lasers have been used to investigate many types of nonlinear dynamic processes [17–19]. In our opinion, it would be useful to investigate a few coupled oscillators system [20, 21] with the help of a multicavity system

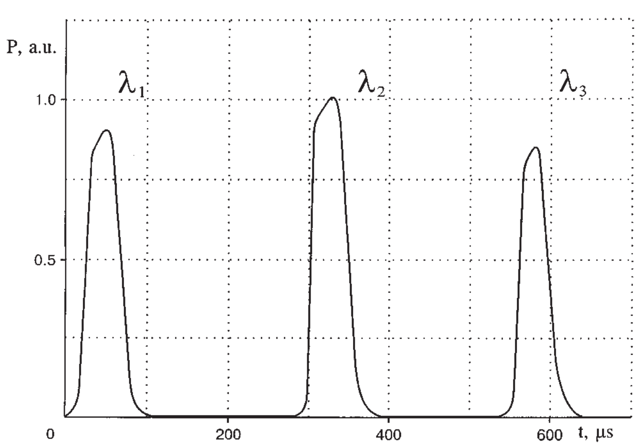

Fig. 4. A series of pulses temporally separated with the help of rotatable diffraction grating: $\lambda_1 = 5.389\,\mu m$ (9-P(20)), $\lambda_2 = 5.809\,\mu m$ (15-P(16)), $\lambda_3 = 6.178\,\mu m$ (19-P(17))

coupled through the common active medium. In this case the coupling efficiency can be easily varied from low (lines belong to the different vibrational bands) to high (lines belong to one vibrational band).

4 Conclusions

In this work multiline oscillation on several transitions in a cw CO laser has been investigated by use of a multichannel cavity. The field-mechanism approach was applied for experimental data analysis. The criteria for selection of the optimal lines for stable multiline oscillation were determined. A stabilized three-color CO laser developed on the basis of these investigations has been utilized for the nondestructive testing of the quality of industrial silicon ingots.

The field-mechanism approach may be useful when considering simultaneous multiline oscillation in other molecular gas lasers (e.g., N_2O, HF, DF, etc.). It should be noted that in the case of simultaneous oscillation of two orthogonally polarized waves on the same transition [5], to weaken the competition influence one needs to switch on the polarization bottleneck effect, which is defined by the orientational constant of the transition dipole moment.

It is also possible to use our approach to investigate simultaneous oscillation on different molecular components in the same active medium (CO-CO_2, CO_2-N_2O, $^{12}CO_2$-$^{13}CO_2$) where a common reservoir of stored vibrational energy is used jointly [22, 23]. In this case the bottleneck effect is determined with an intermolecular exchange. Preliminary experiments with a cw CO-CO_2 laser which has an output containing 5.4 and 10.6 μm radiation confirm this conclusion.

Acknowledgements. The authors wish to express much appreciation and thanks to Dr. V.V. Litvinov for much assistance with the work and discussions and Dr. V.N. Chizhenvsky for his interesting idea and helpful discussion of nonlinear dynamics. We are also grateful to Dr. R. Williams (Oxford University) for his assistance in editing the text. This work was partly supported by a grant from the Mayer fund, which has been awarded by the American Physical Society, and by the Fundamental Research Foundation of the Republic of Belarus (grant F95-208).

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