An optically pumped carbon monoxide laser operating at elevated temperatures

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Received 20 May 2013, in final form 8 July 2013
Accepted for publication 22 July 2013
Published 19 August 2013
Online at stacks.iop.org/LP/23/095004

Abstract
A flowing gas, optically pumped, CO laser has been designed and built. The laser has been made to operate on the fundamental (≈ 5 µm) infrared bands of the CO vibrational states. The laser is powered by absorption of continuous wave radiation from an electric-discharge-excited CO laser. With this system, the kinetics of the establishment and maintenance of strong population inversions in CO at temperatures above 300 K is studied, independently of the complications of the electron impact processes and of other chemical channels which are present in electric discharge CO lasers. Lasing is obtained at temperatures up to 450 K, well above the cryogenic operating temperatures of conventional electric discharge CO lasers. The vibrational population distribution in the optically pumped laser is measured and the laser output power is determined as a function of the system operating parameters. Laser power conversion factors up to 14% have been observed. An optically pumped CO laser kinetic model is used to analyze the experimental results, providing insight into the details of secondary lasing kinetics.

1. Introduction

Carbon monoxide (CO) lasers, operating on the fundamental and first overtone vibrational–rotational bands of CO, are among the most powerful and efficient gas lasers ever developed. Since their first development by Patel [1] and by Osgood and Eppers [2], the highest performance has been achieved in gas systems excited by various electric discharge methods and with the gas cooled to cryogenic temperatures. Efficiencies as high as 50% were reported by Grigor’yan et al [3] and powers up to 200 kW have been obtained by Dymshits et al [4] in cw systems operating on the fundamental bands. Overtone cw lasing was also achieved in similar systems by Bergman and Rich [5], with efficiencies up to 5% reported by Utkin et al [6]. Many of these systems have recently been reviewed in detail by Ionin [7, 8].

The mechanism creating the population inversion in these lasers was first described by Rich [9]. The lower quantum levels of the vibrational mode of CO are excited by inelastic collisions with the discharge electrons. Energy is then rapidly redistributed among the vibrational states by exchange of vibrational quanta in CO–CO inelastic collisions (so-called vibration-to-vibration, or ‘V–V’ exchange collisions). When the mean energy in the vibrational mode exceeds the mean energy in the ‘external’ modes of molecular translation and rotation, an extremely non-Boltzmann distribution of population is maintained among the CO vibrational levels, in accordance with the basic theory of Treanor et al [10]. The distribution is characterized by significant overpopulation of the higher vibrational quantum levels; in some cases, even total population inversions between levels are possible. These inversions are enhanced by either increasing the energy stored in vibration, or by decreasing the energy in the molecular translational and rotational modes, i.e. by decreasing the gas kinetic temperature.

In practice, the powerful electrically excited CO lasers mentioned above use a glow-type electric discharge to provide the vibrational excitation source, and rely on either...
wall-cooling or supersonic expansion to cool the discharge gases. Recently, however, theoretical reactive scattering cross section calculations by Schatz [11] have indicated that collisions of O$_2$ with carbon (C, C$_2$) can generate large amounts of CO in excited internal molecular states. A considerable amount of the reaction energy can appear in CO vibrational mode (up to 50% of the exothermicity of the reaction). These findings raise the possibility of obtaining steady-state vibrational mode power loadings of CO which can exceed those achievable by glow discharge excitation, resulting in efficient CO laser performance. A possible drawback of this approach is that higher gas temperatures will be produced, which can have an opposite effect on performance by reducing the laser gain. The purpose of the present paper is examining the lasing potential of this new method of producing vibrationally excited CO. For this purpose, a conventional CO laser is focused into a flowing gas, optically pumped cell to produce high vibrational mode power loadings. This approach is different from the one used by Anan’ev et al [12], who were the first to use a combination of a pulsed electric discharge in a CO–N$_2$ mixture in an absorption cell and a pulsed CO laser producing additional optical pumping of the discharge-excited mixture in the cell for pump laser energy conversion. In their experiments, discharge excitation was necessary to populate CO vibrational levels by electron impact and enable pump CO laser radiation absorption. Without the discharge, pump laser radiation propagating through the cell (lasing on CO fundamental bands down to $\nu = 4 \rightarrow 3$) was not absorbed. In [12], significant amplification of pump laser radiation by CO excited in the absorption cell was observed on high fundamental bands, up to the 16 $\rightarrow$ 15.

The setup used in the present work is similar to that of Schulz et al [13], who were the first to achieve lasing by optical pumping of CO without the use of auxiliary electric discharge excitation. Unlike the work of Schulz, in the present research, the cell gases can be at elevated temperatures, well above those used in conventional cryogenically-cooled CO lasers. In parallel with the experiment, an optically pumped CO laser kinetic model is used to analyze the experimental results, providing insight into the details of secondary lasing kinetics.

2. Experimental setup

A schematic of the experiment is shown in figure 1. An electric-discharge-excited cw CO laser (‘pump laser’, at the top of the figure) is used to excite the vibrational mode of CO flowing in one of two absorption cells, shown below the pump laser. The laser beam is directed coaxially into the absorption cell and is focused in the center of the cell with a 90 cm focal length lens. In the present experiments, two types of absorption cells have been used. The first cell (cell A in figure 1) is a 1.5 cm diameter $\times$ 100 cm long glass tube with provisions for wall-cooling and inert gas purges at the tube ends. The cooling jacket surrounding the cell tube can be filled with liquid nitrogen, thus providing efficient wall-cooling of the cell gases to cryogenic temperatures. However, in most of present experiments, the cell was not actively cooled. A 1 in diameter IR transmitting CaF$_2$ window in the arm midway between the ends of the tube (see figure 1) provides optical access to the optically pumped CO in the cell, permitting use of FTIR emission spectroscopy (Bruker IFS-66 spectrometer, InSb detector) to measure CO vibrational populations and the translational/rotational temperature, which is equivalent to the gas kinetic temperature. A long-pass filter (cutoff near 4.88 $\mu$m) was used to prevent aliasing in the CO emission spectra.

For the measurements reported here, a strongly vibrationally pumped CO region extends for 30–100 cm along cell A, varying with operating conditions. A gas mixture
flowing through the cell (at flow velocity of several m s\(^{-1}\)), is 3–10% CO in Ar, at a total gas pressure of \(P = 10\) Torr. Brewster windows are employed at the tube ends to reduce intracavity losses. A laser resonator optical cavity is established in the cell using a custom-order dichroic mirror (Rocky Mountain Instruments) at one end, and a near total-reflection, silver-coated mirror at the other end. The dichroic mirror allows most of the short-wavelength radiation from the pump laser, which is absorbed by the lower CO vibrational states \((v = 0 \text{ to } v \sim 10)\), to enter the cell. The dichroic mirror rejects most longer-wavelength radiation coming from vibrational transitions originating in levels \(v \sim 11\) and higher. The intracavity laser field is sampled using the partial reflection from a Brewster window (see figure 1), which is directed to a second FT spectrometer (Varian 660 IR, HgCdTe detector). Spectra of the laser radiation within the optically pumped cavity are obtained and analyzed by this instrument.

The second cell (cell B in figure 1) is a 5 cm diameter \(\times\) 100 cm long glass tube equipped with multiple CaF\(_2\) optical access windows as well as with inert gas purges at the tube ends. A CO–Ar mixture is slowly flowing through the cell (1–2% CO in the mixture, total pressure \(P = 10\) Torr, flow velocity several cm s\(^{-1}\)), and argon is injected into the purges. The distance between the ends of the purges is 50 cm. The main purpose of using this larger diameter cell is to sustain optically pumped CO vibrational distributions in a slowly flowing CO–Ar mixture at relatively high temperatures, up to \(T \sim 500\) K, without generating significant carbon deposits on the walls of the tube and overheating them, as well as to provide better optical access. Similar to cell A, Brewster windows are attached to the ends of the cell. The same dichroic ‘output coupler’ mirror and same ‘total reflector’ mirror are used in the optical resonator. Infrared emission spectra of optically pumped CO are measured by a Bruker IFS-66 spectrometer, while pump laser and secondary lasering spectra originating in the optically pumped cell are measured by a Varian 660 IR spectrometer (see figure 1). The pump laser/secondary laser beams are reflected off a CaF\(_2\) plate into the spectrometer to reduce the intensity, as shown in figure 1.

The pump laser power transmitted into the resonator through the dichroic mirror (approximately 60% of the incident power), as well as the sum of pump and secondary laser power reflected from the CaF\(_2\) plate, are measured by Scientech Vector S310 and Coherent Ultima power meters. The laser power conversion ratio, i.e. the ratio of the secondary laser power to the pump laser power after the dichroic mirror, is determined from these two measurements using the known CaF\(_2\) plate reflection coefficient (in the range of several per cent), which is measured separately without moving the plate. This approach was employed to measure the power conversion ratio obtained both in cell A and in cell B.

3. Modeling and analysis

The kinetic model of the optically pumped CO laser incorporates kinetic equations governing the population of each vibrational quantum level for each diatomic species in the laser gas mixture (at the present conditions, only CO) as a function of time (‘master equation’ modeling). Coupled with these equations is a set of laser field equations governing the stimulated emission intensity on each lasing vibrational transition as a function of time. The laser cavity model is a simple Fabry–Perot model. On each vibrational transition, lasing is assumed to occur on the vibrational–rotational transition with maximum gain. In the baseline version of the model, spatial homogeneity of the cell gases, pump laser intensity distribution, and the stimulated emission radiative field is assumed. Input parameters specified are the pump laser power and spectrum, estimated pump laser beam focal area used as an adjustable parameter, gas species concentrations, the gas translational/rotational mode temperature, the cavity mirror reflectivities, cavity gain length, and cavity optical losses. The code calculates the laser output on each vibrational transition as a function of time. If a constant pump laser radiation field is specified, the equations are integrated to steady state, resulting in the prediction of the cw laser output from the cell.

The model is based on the OSU CO optically pumped plasma model [14] and the CO laser field model of Rockwood and Brau [15]. The best available state-to-state vibrational energy transfer rates for CO are used, as discussed in detail in [14]; some of these are supplied by recent OSU theoretical and experimental results [14, 16–19]. Fundamental and overtone radiative transitions are incorporated. The model also includes the rapid collisional energy transfer from ground electronic state (\(X^3\Pi\)) vibrational levels to the \(A^1\Sigma^+\) excited electronic state, as observed in our previous measurements [20]. The CO laser model was validated by coupling the optical pumping model with Boltzmann equation for plasma electrons and incorporating CO vibrational excitation processes by electron impact, and comparing its predictions with the output spectra of fundamental and first overtone band electric discharge CO lasers [3, 6, 14]. The results of the validation showed that the calculated and experimental laser spectra agree quite well.

To estimate the effect of radial laser power distribution, radial diffusion, and heat conduction in CO–Ar optically pumped by a focused laser beam, as well that of spatial integration of CO emission by the FTIR spectrometer, we used an expanded version of the kinetic model of optically pumped CO incorporating these effects [14]. In addition to master equation for CO vibrational populations, the expanded model incorporates a Gaussian laser power distribution, diffusion of vibrationally excited molecules out of the laser beam, energy equation predicting radial temperature distribution in the slowly flowing optically pumped plasma cooled by diffusion and conduction heat transfer, and line-of-sight integration of CO emission by the spectrometer. In these calculations, the upper bound of the pump laser beam waist was estimated from the diameter of the high-temperature luminous spot created by the focused laser beam on a beam stop.

4. Results

In this section, results of CO optical pumping/secondary lasing experiments are presented for three representative
Figure 2. CO infrared emission spectra from the optically pumped cell, showing first and second overtone, as well as a portion of the fundamental. 2% CO in Ar, $P = 10$ Torr. Translational–rotational temperature inferred from the fundamental spectrum $T = 415$ K. The two spectra shown are taken with the optically pumped cell resonator in and out of alignment.

cases. Case I and II results were obtained in the first, relatively fast flow absorption cell (cell A in figure 1), with 3% CO and 10% CO in Ar, respectively. Case III results were obtained in the second, slow flow cell (cell B in figure 1), with 2% CO in Ar. For all three cases, the pressure in the cell was kept the same, $P = 10$ Torr, and pump laser power was in the range 14–15 W, of which approximately 60% (i.e. 8.8–9.5 W) was transmitted into the cell resonator through the dichroic mirror. Although cell A allows the use of external wall-cooling by liquid nitrogen, no cooling was used in any of these cases.

Figure 2 shows typical CO infrared emission spectra from the optically pumped cell (cell B), taken through one of the optical access windows in the side of the cell (see figure 1). The cell operating parameters for the case of these spectra are 2% CO in Ar, at a total pressure of 10 Torr and a flow velocity of approximately 24 cm s$^{-1}$. Under these conditions, the optically pumped region of vibrationally excited CO is approximately 1 cm in diameter and extends for approximately 50 cm along the cell, occupying nearly the entire distance between the purges (see figure 1), as evidenced by a visible blue glow radiated on the Swan bands of C$_2$ produced by chemically reacting, highly vibrationally excited CO [21]. The structures above 4300 cm$^{-1}$ are the second overtone bands of CO; the intense structure between 4300 cm$^{-1}$ and 2200 cm$^{-1}$ is primarily due to the first overtone bands of CO; the large structure at 2200 cm$^{-1}$ is the beginning of the R-branch of the fundamental band. These lines are quite intense and well-resolved. They are used to infer the rotational–translational temperature, i.e., the gas kinetic temperature. The temperature inferred from the spectra shown in figure 2 is $415 \pm 10$ K.

Figure 3 plots relative populations of CO vibrational levels inferred from emission spectra such as those shown in figure 2, at the conditions of case I (cell A in figure 1, 3% CO in Ar, $P = 10$ Torr, $T = 335 \pm 10$ K), plotted against vibrational quantum number. These populations are inferred from two spectra such as shown in figure 2, as discussed by Ploenjes et al [14]. The populations inferred from the FTIR emission spectra are represented by filled circles. Note that vibrational quantum levels above $v = 40$ are not populated. This is caused by rapid energy transfer from the ground electronic state ($X^1\Sigma_g$) vibrational levels to the $A^1\Pi$ excited electronic state [20]. This energy transfer process provides a major energy sink and stops the further up-pumping of higher vibrational levels. It can be seen that the vibrational populations with the resonator in alignment are greatly reduced from those of the misaligned case, beginning...
at $v \sim 10$. This is because, when well aligned, the optically pumped cell is lasing, which depopulates the lasing levels, and substantial power is being removed by the laser beam passing through the dichroic mirror serving as an output coupler (see figure 1). The effect of this removal of the excited CO energy is also apparent from visible observation of the cell. As the mirrors are moved into or out of alignment, the visible emission from the cell ($C_2$ Swan bands), arising primarily from $C_2$ generated in $CO + CO$ chemical reactions [21], strikingly diminishes as laser power is extracted. Most importantly, the gas kinetic temperature of $T = 335$ K is fairly high compared to that which typically produces such large population inversions among the vibrational levels.

In figure 3, the solid lines are the vibrational populations predicted by the kinetic model for the cell conditions stated on the figure, with a pump laser power of 9.4 W (measured after the dichroic mirror) admitted into the cell. It can be seen that the effect of depopulating high vibrational levels by stimulated emission when the resonator is brought into alignment is reproduced well by the model. Further results of theoretical modeling calculations for the conditions of the experiment reported in figure 3 are given in figure 4. The predicted small signal gain for the highest gain rotational line on each fundamental band vibrational transition (left axis) is plotted for the highly pumped distribution in figure 3, i.e., the unaligned case, with no laser power being extracted. Highest gain, 0.145% cm$^{-1}$, is predicted on $v = 15 \rightarrow 14$ vibrational transition. The vibrational–rotational transition predicted to have the highest small signal gain is also shown (right axis). It can be seen that most of the highest gain lines are for $J'' = 15$ ($P(15)$), the relatively high rotational quantum number reflecting the relatively high temperature in the optically pumped cell. Additional modeling results for these cell conditions are summarized in figure 5. The experimentally measured intensities of the pump laser lines entering the cell are shown in blue on the left of the spectrum. The blue triangles are the measured identifications ($J$ values) of these lines. It can be seen that these lines range from $P(7)$ to $P(13)$, the low $J$ values being characteristic of the cryogenic temperature ($T \sim 150$ K) of the liquid nitrogen cooled pump laser. The model-predicted laser power spectrum from the aligned resonator in the cell is shown in red on the right. It can be seen that the theoretical model predicts substantial lasing from the cell despite the high temperature of the optically pumped mixture. Indeed, the predicted conversion ratio (secondary laser power out to pump laser power transmitted into resonator) is 28% for this case. This prediction assumes single-pass gain length of $L = 100$ cm and no resonator losses except through the output coupler dichroic mirror. Note that the predicted cell lasing lines are in the range $P(16)$–$P(19)$, higher rotational lines than those of the pump laser ($P(8)$–$P(13)$), reflecting the higher temperature in the cell. These results suggest a simple way of differentiating between the pump laser lines and the secondary lasing lines, based on their widely different $J$ values.

Intracavity laser field spectra acquired at the same conditions as in figure 3, are shown in figure 6. The lower part of the figure shows the laser spectrum with the cavity aligned, but with no CO in the cell. All that appears is the pump laser radiation. The upper part of the figure shows the intracavity field with 3% CO added, at the conditions of figure 3. Also shown (black curve) is the reflectivity of the dichroic mirror. The reflectivity is designed to enhance laser radiation from upper vibrational levels, below 1925 cm$^{-1}$. 

![Figure 4](https://example.com/figure4.png)

**Figure 4.** Case I. Small signal gain predicted by the kinetic model at the conditions of figure 3 (resonator out of alignment).

![Figure 5](https://example.com/figure5.png)

**Figure 5.** Case I. Pump CO laser spectrum (blue) measured after the dichroic mirror (9.4 W) and optically pumped laser spectrum (red) predicted by the model at the conditions of figure 3 (resonator in alignment). Predicted conversion efficiency 28%.
These levels are not directly coupled to the pump radiation, but are populated by the V–V up-pump process. It can be seen that the spectrum on the top shows amplification of some of the pump laser radiation transitions, but there are also new laser lines for bands below 1910 cm\(^{-1}\), originating from the higher vibrational levels. This is clear proof that the cell is lasing on the upper vibrational bands, as predicted by the kinetic model (see figure 5). The pump laser transitions are plotted in blue while the secondary laser transitions, identified by their high \(J\) values, are shown in red. Laser output from the pump cell is created on the \(v = 7 \rightarrow 6\) band up to the \(v = 15 \rightarrow 14\) band. These vibrational–rotational lines are not present at all in the pump laser spectrum. It is also important to note that the rotational levels lasing, as indicated on the figure, are states \(J'' = 15–23\). These are relatively high rotational states, corresponding to the high gas temperature in the cell \((T = 335\, K)\). In contrast, the pump laser radiation, coming from the liquid nitrogen cooled CO pump laser, originates from the rotational states \(J'' = 12\) and lower, corresponding to the low gas temperature in the cooled laser, \(T \sim 150\, K\).

These experimental findings are consistent with the lasing predictions of the model, as given in figure 5. On the other hand, detailed comparison of figures 5 and 6 shows that secondary lasing is produced on somewhat lower vibrational bands, down to \(v = 7 \rightarrow 6\) (the lowest secondary lasing band predicted by the model is \(v = 9 \rightarrow 8\), see figure 5), and on somewhat higher \(J\) values (up to \(J'' = 23\) compared to \(J'' = 18\) predicted by the model). This suggests that both populations of vibrational levels \(v \sim 7–8\) and rotational temperature in the high-gain region in the resonator (most likely on the laser beam centerline) are somewhat higher than the model predicts. This issue will be discussed in greater detail at the end of this section. It can also be seen that the model predicts lasing on higher bands, up to \(v = 23 \rightarrow 22\) (see figure 5), which are not detected in the experimental laser spectra (see figure 6). This is most likely due to additional resonator losses, set to zero in the present modeling calculations. Finally, laser power conversion ratio measured at these conditions, 6.7–14.4% depending on the resonator alignment, is lower than predicted by the model, 28%. Power conversion ratio may be reduced both by the resonator losses and by a shorter effective gain length in the cavity, controlled by the length of the optically pumped region, which may well be shorter due to pump laser beam absorption along the cell.

Further tests were conducted to confirm that the observed laser lines originating from high vibrational levels and high rotational levels are indeed generated by the population inversions in the optically pumped cell. Intracavity laser field spectra were acquired for the conditions of figure 3, and figure 7 displays these for the cell resonator in alignment (top), and for the resonator deliberately out of alignment (bottom). The additional laser lines generated within the cell, shown in red, are apparent below 1910 cm\(^{-1}\). Again, it is obvious that nearly all the upper-band lasing arises from the optically pumped cell.

The results for case II conditions (10% CO in Ar, \(P = 10\,\text{Torr},\, T = 360 \pm 10\,\text{K}\)) are summarized in figures 8 and 9. In this case, the vibrationally pumped region visibly extends for only approximately 30 cm along the cell, most likely due to more significant absorption of the pump laser radiation compared to conditions of case I (3% CO in Ar). Figure 8 shows the distribution of vibrational level populations as measured by FTIR emission spectroscopy. Again, the populations are shown as the red filled circles for the resonator out of alignment (no laser power being extracted), and the blue filled circles for the resonator in alignment (laser energy being removed). The corresponding vibrational distribution functions calculated by the kinetic modeling code for these cases are plotted as the solid lines. Again, significant reduction in populations accompanying removal of energy by lasing is apparent. The intracavity laser field spectra for these conditions are given in figure 9. The top and bottom parts of every plot in the figure show,
as before, a comparison between the spectra with the cell cavity misaligned and aligned. With the cell unaligned, a few pump laser lines are passed into the cell and are not totally absorbed by the cell CO. In contrast, with the cell resonator aligned, there are a large number of intense lasing transitions arising from the vibrationally pumped cell gases. These lasing transitions extend to very high vibrational levels. The inset of figure 9 is an enlargement of the long wavelength portion of the spectrum; the individual lasing vibrational–rotational lines are identified above each line. Here, lasing from the cell is observed on the higher vibrational transitions $v = 8 \rightarrow 7$ up to $v = 20 \rightarrow 19$. The lasing rotational states are in the range from P(16) to P(22). The high rotational quantum levels reflect the increased populations of such states at the cell temperature of $T = 380$ K. The observed lasing ro-vibrational levels are consistent with the model predictions, although, similar to case I, lasing occurs on lower vibrational transitions compared to the model predictions (down to $v = 8 \rightarrow 7$ versus $v = 11 \rightarrow 10$) and up to higher rotational transitions (P(22) versus P(20)). Again, this suggests that populations of vibrational levels $v = 8-10$ and rotational temperature in the high-gain region in the resonator (on the laser beam centerline) are higher than the model predicts. Laser power conversion ratio measured at these conditions, 2.7%, is significantly lower than predicted by the model, 18%, indicating that gain length in the cavity is considerably shorter than $L = 100$ cm assumed in the modeling calculations.

Optical pumping and secondary lasing in cell A has been also achieved at cryogenic temperatures, maintained by wall-cooling the cell with a liquid nitrogen bath. At these conditions, the measured temperature in the cell was $T = 155 \pm 5$ K. The same qualitative behavior has been observed as in cases without wall-cooling (cases I and II), but with the secondary lasing transitions produced at lower rotational quantum numbers, P(11) to P(16), similar to the ones in the pump laser, P(9) to P(14), reflecting similar temperatures in the two lasers.

Finally, figures 10 and 11 show case III results obtained in cell B, where the flow was deliberately slowed down to the velocity of a few cm s$^{-1}$, to increase residence time in the cell. This was done to reach higher temperatures in the optically pumped gases, and estimate the highest temperature at which secondary lasing could be detected at the present conditions. Doing this in the smaller diameter cell A would produce significant carbon deposits on the walls, which could result in overheating the walls by the peripheral region of the laser beam. At the flow velocity of 7–24 cm s$^{-1}$, the estimated residence time in the cell was 2–7 s, which is

![Figure 8](image_url)

**Figure 8.** Case II. Symbols: CO vibrational distribution functions measured in the optically pumped 10% CO–Ar mixture at $P = 10$ Torr at $T = 360$ K, with the resonator in and out of alignment. Lines: kinetic model predictions at the same conditions. Reduction of high CO vibrational level populations, caused by stimulated emission in the cell during lasing, is apparent.

![Figure 9](image_url)

**Figure 9.** Case II. Laser (intracavity flux) spectra produced by the optically pumped CO cell with the resonator in alignment and out of alignment (top and bottom parts of each plot). 10% CO in Ar, $P = 10$ Torr, $T = 380$ K. Additional laser lines generated in the optically pumped cell are shown in red. Lasing is observed on vibrational bands up to $v = 20 \rightarrow v = 19$ (P(16) line at $\lambda = 6.258 \mu$m).
much longer than the characteristic time for diffusion at these conditions, \( \sim 0.1 \) s, such that the radial distributions of temperature and vibrational level populations in the cell are controlled mainly by conduction heat transfer and diffusion. Indeed, the apparent diameter of the optically pumped region in cell B, estimated from the size of the \( \mathrm{C}_2 \) Swan emission region, \( \sim 1 \) cm, is significantly larger than that observed in cell A, \( \sim 3\text{–}5 \) mm. Also, \( \mathrm{C}_2 \) Swan glow in cell B extended over the entire length of the cell between the two Ar purges (see figure 1), providing a fairly accurate estimate of the gain path, \( L \approx 50 \) cm.

In case III (2% CO in Ar, \( P = 10 \) Torr, \( T = 415 \pm 10 \) K), the same qualitative behavior is observed as in the lower temperature cases, case I and case II. Specifically, figure 10 shows the measured and predicted vibrational level populations, again with the cell resonator in and out of alignment. At these higher temperature, shorter cavity gain conditions, the reduction in the pumped vibrational level populations with lasing was not nearly as significant as in cases I and II (compare figure 10 with figures 3 and 8), although greater than the uncertainty in the inference of vibrational level populations (2–8% for \( v = 10\text{–}38 \) in figure 10). Small signal gain for this higher temperature case, at the conditions when vibrational populations predicted by the model are close to the populations inferred from CO emission spectra, is significantly lower compared to cases I and II, only 0.027% cm\(^{-1}\) for \( v = 16 \rightarrow 15 \) transition. However, it is obvious that if gain in the resonator were indeed that low, lasing would be impossible. Indeed, lower bound threshold gain calculated for the highest reflectivity of the output coupler dichroic mirror, \( r_1 = 0.94 \), and the reflectivity of the ‘total reflector’ mirror, \( r_2 = 0.99 \), is \( \gamma = -\ln(r_1r_2)/2L = 0.072\% \) cm\(^{-1}\), nearly a factor of 3 higher compared to the predicted value. Note that uncertainties in the temperature in the optically pumped cell, mirror reflectivities, and gain length are much smaller. Since secondary lasing at these conditions has been detected, this suggests that both CO vibrational level populations and gain on the laser beam centerline are in fact significantly higher than predicted by the model.

As an estimate of how much the model may underpredict gain on the centerline, secondary lasing spectrum was calculated for single-pass gain length of \( L = 200 \) cm, i.e. by a factor of 4 longer than the distance between the Ar purges (see figure 1). For these conditions, the model predicts laser power conversion coefficient of 8.0%, closer to the experimentally measured value at these conditions, 14.4%. This demonstrates that peak gain is underestimated by the model by at least a factor of 4. The experimental laser spectra measured outside of the resonator, with and without CO in the absorption cell, are shown in figure 11. In both figures, the spectrum at the top is with 3% CO in the cell and the spectrum at the bottom is without CO in the cell, which merely reflects the unabsorbed radiation from the pump laser. Again, additional laser lines are generated by the optically pumped cell. Similar to cases I and II, additional lines and higher lasing vibrational levels generated by the cell up-pumping are apparent. Note that spectra shown for cases I and II (figures 6, 7 and 9, respectively) are measured intracavity, while spectra shown for case III (figure 11) are measured outside of the resonator.

To quantify the effect of the temperature in the optically pumped cell on the secondary lasing, temperature, CO vibrational populations, and laser power conversion ratio were measured at case III conditions (3% CO in Ar, \( P = 10 \) Torr), for three different CO–Ar flow velocities in the cell, 24, 11, and 7 cm s\(^{-1}\). The results are summarized in table 1.

These results demonstrate that high laser power conversion can be achieved in the optically pumped CO laser operated without cooling, at temperatures significantly exceeding room temperature, up to at least \( T = 450 \) K. Further increase in the conversion coefficient may be achieved by
Table 1. Absorption cell operating conditions.

<table>
<thead>
<tr>
<th>Cell conditions (case III)</th>
<th>Flow velocity (cm s(^{-1}))</th>
<th>Temperature (K)</th>
<th>Power conversion coefficient (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3% CO, ( P = 10 ) Torr</td>
<td>24</td>
<td>415</td>
<td>14.4</td>
</tr>
<tr>
<td>3% CO, ( P = 10 ) Torr</td>
<td>11</td>
<td>450</td>
<td>8.3</td>
</tr>
<tr>
<td>3% CO, ( P = 10 ) Torr</td>
<td>7</td>
<td>470</td>
<td>No lasing</td>
</tr>
</tbody>
</table>

operating the pump laser in a repetitively pulsed (Q-switched) mode, using an intracavity chopper or a rotating mirror. Pulsed mode operation could produce high transient CO vibrational level populations, without significant increase in temperature, resulting in an even higher laser power conversion factor.

As discussed in section 3, we used an expanded kinetic model of optically pumped CO mixtures excited by a focused laser beam [14] to estimate the effect of radial laser power distribution and diffusion on laser gain, as well as the effect of spatial integration of CO emission spectra by the FTIR spectrometer on inferred vibrational level populations. In these calculations, the upper bound of the pump laser beam waist was estimated from the diameter of the spot created by the focused laser beam on a brick used as a beam stop, \( \approx 4 \) mm. Assuming the radius of the Gaussian beam where the power density decreases by a factor of \( 1/e \) to be 2 mm gives beam FWHM of 2.8 mm. In the calculations, a CO–Ar mixture, initially in equilibrium, was excited by the focused laser beam until a steady-state CO vibrational populations distribution and temperature distribution were achieved across the entire cross section of the cell (radius of \( r = 25 \) mm). The results are summarized in figures 12–15. From figure 12, it can be seen that strong vibrational nonequilibrium is achieved both in the region excited by resonance absorption of the laser beam and outside of the laser beam, where nonequilibrium is maintained by radial diffusion of vibrationally excited, slowly relaxing CO molecules. As expected, both CO ‘first level’ vibrational temperature, \( T_v = [E_{v=1} - E_{v=0}]/\ln(n_0/n_1) \), and the gas temperature peak on the beam centerline (see figure 12), CO vibrational level populations, also peaking on the centerline (at \( r = 0 \)), gradually decrease along the radial coordinate (see figure 13). As can be seen from figure 13, the line-of-sight integrated vibrational populations, which are in good agreement with the populations inferred from the FTIR emission spectra, are considerably lower than the populations on the centerline, by a factor of 2–5.

Figure 14 compares small signal gain at different radial locations in the cell with the small signal gain predicted by a ‘zero-dimensional’ kinetic model used to predict vibrational level populations, gain, and secondary laser spectra in figures 3–5 (case I), figure 8 (case II), and figure 10 (case III). It can be seen that peak gain on the centerline exceeds the value predicted by the 0D model by approximately a factor of 2.5. Moreover, gain in the entire region directly excited by the
Figure 14. Comparison of small signal gain at different radial locations predicted by the kinetic model incorporating radial diffusion/line-of-sight emission averaging with the prediction of a 0D kinetic model. Conditions same as in figure 12.

pump laser beam ($r < 5 \text{ mm}$) is significantly higher than gain predicted by the 0D model. Finally, figure 15 compares CO vibrational distribution functions predicted by the two models (0D and 1D line-of-sight integrated) as well as gain predicted by the two models (0D and 1D on the centerline). Note that the distribution functions predicted by both models are close to each other and in good agreement with the populations inferred from the emission spectra. However, as mentioned before, gain on the centerline is considerably higher compared to the value predicted by 0D model, by about a factor of 3 at $v \sim 15$ and by an order of magnitude at $v \sim 10$. Also, high gain on the centerline extends to much lower CO vibrational transitions compared to the predictions of the 0D model, which may explain why secondary lasing in cases I–III is consistently detected on lower vibrational bands compared to the model predictions.

These results demonstrate that line-of-sight integration of CO emission by the FTIR spectrometer yields spatially averaged CO vibrational populations which are in fact significantly lower than the populations in the strongly excited region near the centerline of the pump laser beam. Basically, matching these averaged populations by a 0D kinetic model considerably underestimates peak gain in the absorption cell optically pumped by a focused laser beam. This effect becomes far more pronounced as the cell diameter is increased and the flow velocity is reduced, such that the diameter of nonequilibrium optically pumped region is enhanced considerably by radial diffusion, as occurs in cell B. At these conditions, the relative contribution of the regions on the periphery of the cell into the emission signal becomes especially significant compared to the signal originating near the centerline. The results of the modeling calculations shown in figures 12–15 are strongly dependent on the pump laser beam diameter as well as on relative contributions of different regions along the line-of-sight of the spectrometer into the emission signal, which are subject to significant uncertainty. Therefore the modified model cannot be expected to yield accurate quantitative predictions of radial distribution of gain in the optically pumped cavity. However, we believe that the fact that secondary lasing is detected at the conditions when ‘apparent’ 0D small signal gain is far below lasing threshold (case III) is a clear indication that CO vibrational level populations and gain on the beam centerline are significantly higher. Calculations using the expanded kinetic model support this conclusion.

5. Summary

It is important to note that cw lasing by an optical pumping arrangement similar to that of the present paper has been previously achieved by Schulz et al [13]. However, this result was obtained at cryogenic temperatures by wall-cooling of their cell and there was no direct measurement of the cell gas temperature or measurement of the vibrational level populations. We assume that in [13] the temperature was similar to that measured for the cooled case, $T = 155 \pm 5 \text{ K}$, reported in the present paper. The work of Schulz et al is the only other result that reports lasing from optically excited, V–V pumped CO, without the use of electric discharge excitation. The work of Anan’ev et al [12], where the pulsed pump CO laser energy conversion/amplification in an absorption cell was reported, required initial vibrational excitation of CO in the cell by a pulsed electric discharge. The optically pumped CO experiments reported by McCord et al...
[22, 23] differ significantly from the present study. In [22, 23], the lowest two vibrational levels of CO were pumped by a pulsed tunable source, and pulsed lasing action from CO on these levels was achieved; the system acted as a frequency converter, with no effort to obtain high vibrational level power loadings, which is a major goal of the present research. The present results are the first demonstration of cw lasing action created by vibration–vibration up-pumping in CO at high temperatures. We conclude that it is possible to create an efficient optically pumped CO laser by extreme power loading of the vibrational mode, even at temperatures considerably above room temperature.

Acknowledgments

The support of the Lockheed Martin Company is gratefully acknowledged. Support for Dr Ivanov has been from the Chaszeyka Grant to the Nonequilibrium Thermodynamics Laboratories.

References