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Measurement of Rotational Temperature in CO_2 Waveguide Laser Medium

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Abstract. A small-signal gain in CO_2 waveguide laser medium has been measured on rotational-vibrational transitions in the P-branch of the $(0,0,1)$–$(0,2^0,0)$ band. It has been found that the rotational temperature is well defined in the waveguide laser system where high excitation power is injected and a large amount of energy is flowing through vibrational, rotational, and translational degrees of freedom. The rotational temperature is slightly higher than the translational temperature.

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CO_2 lasers may be classified into three typical types; an ordinary CO_2 laser at low gas pressure with low excitation, a TEA laser at high gas pressure with high excitation, and a waveguide laser at relatively low gas pressure with high excitation. A laser can be described as a stationary nonequilibrium system connected to two thermal reservoirs with different temperatures (pumping source at high temperature and heat sink at low temperature). In the case of CO_2 laser medium, most of energy is first supplied to the vibrational states, then transferred to the rotational and translational degrees of freedom, and it is finally dissipated into a coolant. A fraction of the supplied energy is emitted as a coherent radiation. It is interesting to investigate under various conditions of the laser gas whether temperature is well defined within individual degrees of freedom or not. Furthermore, if the temperature is clearly determined, the relation among the temperatures of individual degrees of freedom will give information on relaxation processes in a nonequilibrium system.

It has been confirmed that in an ordinary CO_2 laser the vibrational mode-temperatures and the rotational temperature are clearly determined, and the Maxwell-Boltzmann distribution is realized in each degree of freedom [1–3]. Relaxation process within a degree of freedom such as the $V-V$ transfer [4] is fast enough to smooth out the deviation from the Maxwell-Boltzmann distribution which is caused by a level-selective excitation. Several workers have reported that the rotational and vibrational temperatures are well defined also in TEA CO_2 laser medium at high pressure under pulsed discharge [5, 6].

On the other hand, no detailed studies on the temperatures in CO_2 waveguide laser medium have been reported so far. A waveguide laser (WGL) is characterized by high discharge current density (about several tens times as large as that in an ordinary CO_2 laser) and relatively low laser gas pressure (typically $50 \sim 100$ Torr), namely it is characterized by a fast excitation and not so fast relaxation. Therefore it is not evident that the population distribution in each degree of freedom is well described by a single temperature in the WGL medium.

We have measured in a Pyrex waveguide laser a small-signal gain on rotation-vibration transitions of the $(0,0,1)$–$(0,2^0,0)$ band as a function of the injected discharge electric power. The rotational temperature is obtained from the small-signal gain measurement. It is found that the rotational temperature is clearly determined and is slightly higher than the translational temperature in the present WGL system.

1. Experimental

The schematic diagram of gain tube structure of the waveguide laser is shown in Fig. 1. The waveguide is a Pyrex capillary of 2 mm inner diameter and 130 mm long. The external diameter of the capillary is 7 mm. Ethanol (~ 250 K) is circulated outside the waveguide

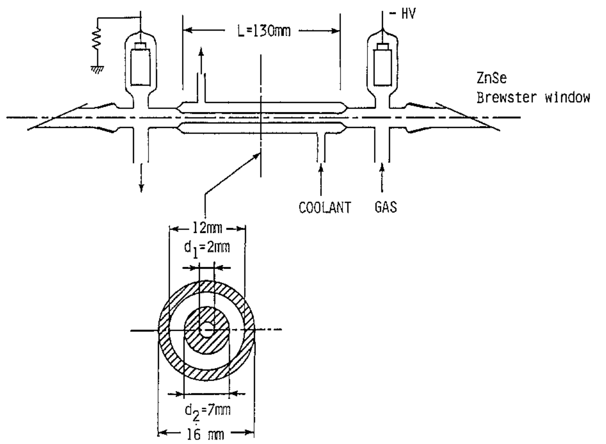

Fig. 1. Schematic diagram of the constructed waveguide laser gain tube. The waveguide is made of Pyrex. The laser tube is sealed by the ZnSe Brewster windows

as a coolant. The laser tube is sealed by two ZnSe windows. A gas mixture of CO_2, N_2, and He (1 : 1 : 4) is flowing through the laser tube. The flow rate is 30 l Torr/s at room temperature. The total gas pressure is 60 Torr at the inlet and 13 Torr at the outlet.

Figure 2 shows the experimental setup to measure the small-signal gain in the waveguide laser tube. The probe light from the single-mode CO_2 laser is introduced into the gain tube. The transmitted output power is monitored by the detector *A* (SAT-A1) and is normalized to the power of the incident light which is detected by the detector *B* (SAT-A1). The wall temperature in the vicinity of the gas outlet is measured with a thermoelectric thermometer to monitor the translational temperature of the laser gas. The discharge current and voltage are measured to obtain the electric power injected to the discharge region.

Figure 3 shows the normalized output power from the detector *A* on the 9 μm P(20) line as a function of the discharge current. The small-signal gain coefficient *g* is derived from the following relation:

$$gL=\ln(Y_I/Y_0), \tag{1}$$

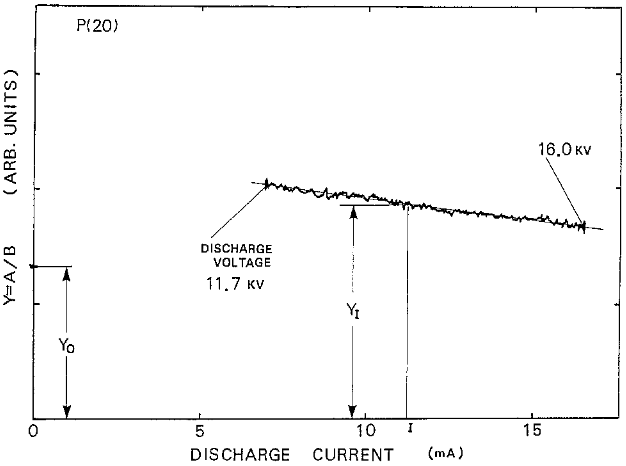

Fig. 3. Normalized transmitted light power as a function of the discharge current. The laser line is 9 μm P(20). The laser gain is decreased monotonically with the current

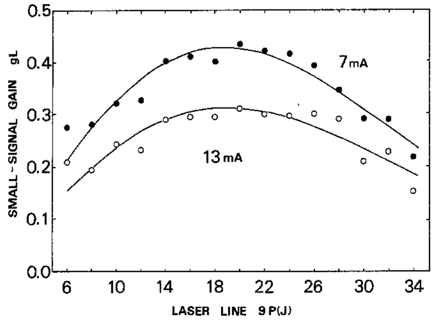

Fig. 4. Gain distribution over the rotational quantum number of the lower laser level *J*. The solid circle represents the small-signal gain at the discharge current of 7 mA, and the open circle at 13 mA. The rotational temperature is determined to be 377 ± 16 K (for 7 mA) and 394 ± 24 K (for 13 mA). The solid line is the best-fit curve of Eq. (2)

where *L* is the waveguide length, and Y_I, Y_0 the normalized output from the detector *A* in the presence and in the absence of the discharge current, respectively. The small-signal gain is plotted as a function of the rotational quantum number *J* in Fig. 4.

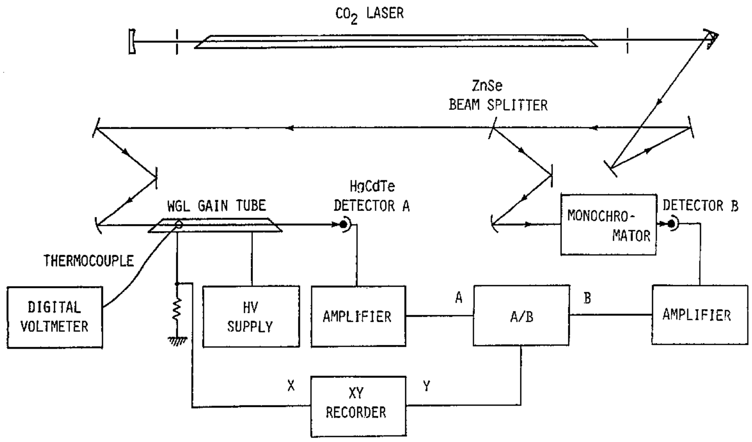

Fig. 2. Experimental setup. The laser beam from the CO_2 laser is split by the ZnSe beam splitter. The transmitted light power is detected by the detector *A* and normalized to the incident light power detected by the detector *B*. A Cu-constantan thermocouple is attached to the surface of the waveguide near the gas outlet, and the wall temperature outside the waveguide is measured with the thermoelectric thermometer

2. Analysis and Discussion

When the rotational temperature and the vibrational temperature of each vibrational mode are well defined, the gain distribution can be written as follows [2, 6]:

$$g(J)L = NJ(R\exp[-B'hcJ(J-1)/kT_r] -\exp[-BhcJ(J+1)/kT_r)], \tag{2}$$

where N is a normalization factor, and B and B' are rotational constants of the (0, 2^0, 0) and (0, 0, 1) vibrational levels, respectively. Here we set $B = B' = 0.39\,\mathrm{cm}^{-1}$ [7]. The rotational temperature is denoted by T_r. The parameter R is expressed as

$$R = \exp(-h\nu_3/kT_3)/\exp(-2h\nu_2/kT_2), \tag{3}$$

where ν_2 and ν_3 are fundamental frequencies, and T_2 and T_3 temperatures of the ν_2 and ν_3 vibrational modes, respectively ($\nu_2/c = 667\,\mathrm{cm}^{-1}$, $\nu_3/c = 2349\,\mathrm{cm}^{-1}$) [7]. The least-squares fitting of Eq. (2) to the observed gain distribution is carried out with the adjustable parameters N, R, and Bhc/kT_r. The fitted curves are shown in Fig. 4 (represented by the solid lines).

The rotational temperature obtained from the fitting is presented in Fig. 5 as a function of the discharge electric power. The error in the determination of the temperature is calculated from the residual of the least-square fitting. It is reasonably small (typically ±20 K), and the rotational temperature is clearly determined in the WGL medium with a high excitation power density. Note that the rotational temperature increases from 375 K to 405 K with the discharge electric power. This heating effect causes a decrease of the laser gain (Fig. 3).

The squares in Fig. 5 represent the wall temperature measured with the thermoelectric thermometer. The difference of the temperature between the wall surface and the laser gas is estimated to be not more than a few degrees by considering the thermal conductivities of the wall and the air outside the wall. The solid line represents the gas temperature theoretically obtained with a model of the heat balance. The calculation well reproduces the absolute values and the dependence of the translational temperature on the discharge electric power. The model is described in detail in Appendix A.

It should be noted that the rotational temperature is systematically higher than the translational temperature beyond the errors. This may be interpreted as follows.

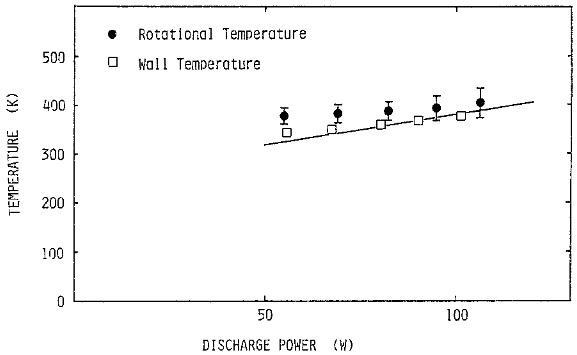

Fig. 5. Dependence of the rotational and translational temperature on the discharge electric power. The circles represent the experimentally obtained rotational temperature. The squares represent the wall temperature measured with the thermoelectric thermometer. The solid line is the theoretical curve calculated based on the model of the heat balance

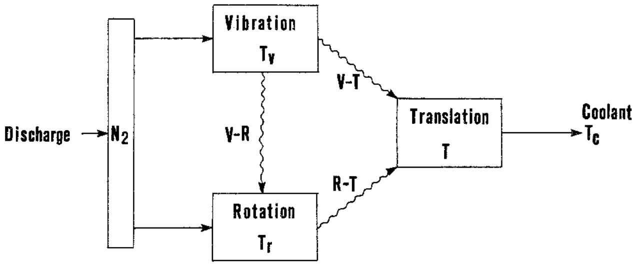

Fig. 6. Schematic diagram of the energy flow in CO_2 laser medium. Discharge electric energy is supplied to the vibrational and rotational degrees of freedom, then transferred through the V–T, V–R, and R–T processes, and finally dissipated into a coolant

In the CO_2 laser medium CO_2 molecules are pumped upon the (0, 0, 1) vibrational state mainly because of the collision with N_2 molecules excited by the electric discharge (see Fig. 6) [7]. The excess vibrational energy is transferred to the rotational and translational degrees of freedom through the V–T and V–R processes [8–10], respectively. The rotational energy is increased also by the collision with N_2 molecules and the electron impact. Energy transfer occurs from the rotational degree of freedom to the translational one. The excitation energy injected from the discharge is finally dissipated to the coolant from the translational degree of freedom.

Thermal equilibrium is realized in each vibrational mode because of the fast V–V processes. The rotational relaxation rapidly smoothes the population distribution distorted by the excitation and the V–R process, and the Maxwell-Boltzmann distribution is maintained also in the rotational states.

The energy ε which is transferred per unit time from the rotational degree of freedom to the translational one can be approximately formulated as

$$\varepsilon = \gamma_{RT}(T_r - T), \tag{4}$$

where T_r and T are the rotational and translational temperatures, respectively, and γ_{RT} the energy flow rate at unit temperature difference $(T_r - T)$. The derivation of Eq. (4) is presented in Appendix B. The factor γ_{RT} increases monotonically as the collisional relaxation rate of the R–T process becomes faster.

In an ordinary CO_2 laser system, a reasonable amount of energy flow is maintained with an undetectably small temperature difference because the relaxation rate of the R–T process is fast enough. However the discharge current density in the present WGL system is about 30 times as large as that in an ordinary CO_2 laser, while the laser gas pressure is not so much higher than that of an ordinary CO_2 laser. A large energy flow is produced by the high excitation, and a detectable temperature difference appears between the rotational and translational degrees of freedom.

3. Conclusion

It has been found from the small-signal gain measurement that the rotational temperature is determined within the error of ±5% in the WGL system. This means that the relaxation rate in the rotational states is fast enough compared with the rate of excitation.

The rotational temperature is equal to the translational temperature in an ordinary CO_2 laser. However, it is obtained to be slightly higher than the translational temperature in the present WGL system. This phenomenon is attributed to a large energy flow due to high excitation power density and relatively low laser gas pressure.

Acknowledgement. The authors gratefully acknowledge Dr. Matsushima for fruitful discussions.

Appendix A

Model of the Heat Balance

A simple model of the heat balance to calculate the translational temperature inside the waveguide is presented here. The schematic diagram of the model is shown in Fig. 7. Consider that the laser gas (flow rate Q mol/s) with the room temperature T^0 comes into the waveguide and gets out of it being heated to the temperature T. The electric power W injected into the discharge region is used to heat the laser gas. The coolant is flowing at the rate Q_c g/s. The coolant is heated from T_c^0 to T_c by receiving thermal energy from the surface of the waveguide.

The following equations are derived from the conservation of energy:

$$Q_cC_cT_c^0+A(T-T_c)=Q_cC_cT_c, \tag{A.1}$$

$$QCT^0+W=QCT+A(T-T_c), \tag{A.2}$$

where

$$A=\frac{2\pi\lambda L}{\ln(d_2/d_1)}. \tag{A.3}$$

Here C is the molar heat of the laser gas, C_c the specific heat of the coolant, λ the thermal conductivity of the wall, and L the waveguide length. The inner and external diameters of the waveguide are denoted by d_1 and d_2, respectively.

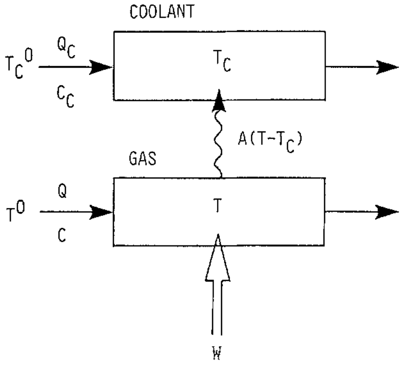

Fig. 7. Schematic diagram of the model to estimate the translational temperature in the waveguide tube

The term $A(T-T_c)$ represents the thermal energy which is transferred from the laser gas to the coolant per unit time. It is derived as follows. Consider a layer with a thickness δr in the wall at a distance r from the waveguide axis. The energy U transferred through the layer is expressed as

$$U=-\lambda\frac{2\pi Lr}{\delta r}\delta T. \tag{A.4}$$

Here δT is difference of the temperature between the inside and outside of the layer. By integrating Eq. (A.4) from the inner radius to the external radius of the waveguide tube, we obtain:

$$U=\frac{2\pi\lambda L}{\ln(d_2/d_1)}(T-T_c)=A(T-T_c). \tag{A.5}$$

From Eqs. (A.1) and (A.2),

$$T=\frac{W+aT_c^0+bT^0}{a+b}, \tag{A.6}$$

where

$$a=\frac{AQ_cC_c}{A+Q_cC_c}, \tag{A.7}$$

$$b=QC. \tag{A.8}$$

The translational temperature T calculated from the model is represented by the solid line in Fig. 5. The parameter values are $\lambda=1.2$ W/m K, $d_1=2$ mm, $d_2=7$ mm, $L=13$ cm, $Q=1.6\times10^{-3}$ mol/s, $Q_c=13$ g/s, $C=13$ J/mol K, $C_c=2.3$ J/g K, $T_c^0=253$ K, and $T^0=295$ K.

Appendix B

Derivation of Eq. (4)

Energy exchange between the rotational and translational degrees of freedom takes place through a process such as

$$CO_2(J+2)+M \rightleftarrows CO_2(J)+M+E_{trans}, \tag{B.1}$$

where a CO_2 molecule in the rotational state with quantum number $J+2$ collides with a collision partner M and makes a transition to the state with quantum number J. The difference of the rotational energy between the initial and final states is transferred to the translational energy of the collision partner E_{trans}. The net energy transferred from the rotational state to the translational state per unit time, ε, is formulated as

$$\varepsilon = \int dE \sum_{J=0}^{\infty} p_J E_{\text{trans}} \times \left(\frac{2J+1}{2J+5}\, n(J+2) m(E) - n(J) m(E+E_{\text{trans}})\right), \tag{B.2}$$

where p_J is the rate of the collision described in Eq. (B.1), $n(J)$ the fraction of CO_2 molecules in the Jth rotational level, and $m(E)$ the fraction of molecules whose translational energy is E, that is,

$$n(J) = Z_{\text{rot}}^{-1}(2J+1)\exp(-BhcJ(J+1)/kT_r), \tag{B.3}$$

$$m(E) = Z_{\text{trans}}^{-1}\exp(-E/kT). \tag{B.4}$$

Here Z_{rot} and Z_{trans} are the rotational and translational partition functions, respectively.

By substituting Eq. (B.3) and Eq. (B.4) into Eq. (B.2) and neglecting the higher order terms of Bhc/kT_r and $(T_r - T)/T$, we obtain,

$$\begin{aligned}\varepsilon &= \sum_{J=0}^{J_{\text{max}}} p_J Bhc(2J+1)(2Bhc(2J+3)/kT)^2 \\ &\quad \times \exp(-Bhc(J+2)(J+3)/kT) T^{-1}(T_r - T) \\ &\equiv \gamma_{\text{RT}}(T)(T_r - T).\end{aligned} \tag{B.5}$$

Here J_{max} is defined as

$$Bhc(J_{\text{max}}+2)(J_{\text{max}}+3)/kT \sim 1. \tag{B.6}$$

It has been shown that the energy transferred through the R–T process is approximately expressed as a product of the difference between the rotational and translational temperature and the coefficient $\gamma_{\text{RT}}(T)$.

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