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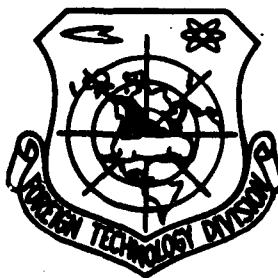


THE VIBRATIONAL RELAXATION PROCESSES IN A $\text{CO}_2\text{-N}_2\text{-H}_2\text{O}$ LASER SYSTEM

by

Yan Haixing

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The Vibrational Relaxation Processes in a $\text{CO}_2\text{-N}_2\text{-H}_2\text{O}$ Laser System

by Yan Haixing
(Institute of Mechanics, Academia Sinica)

Paper received on November 6, 1979

Carbon dioxide Nitrogen
↓
Vibrational relaxation processes and data of a $\text{CO}_2\text{-N}_2\text{-H}_2\text{O}$ laser system up to the present are surveyed. The data before 1988 are based on Taylor & Bitterman's review. The data are treated with weighted least square method to get the best fit in the form of $\log K = A + BX + CX^2 + DX^3$, and the corresponding coefficients are presented. Using these best fits of data and relaxation model with three modes and four vibrational temperatures which is based on the present work, numerical calculation results of small signal gain of GDL agree with experiments, thus eliminating the error introduced by the uncertainties of relaxation data and model.

Chinese language translation

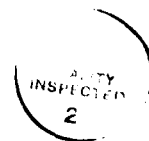
I. Preface

The relaxation process of the vibrational degree-of-freedom inside molecules has a great effect on the fields of high speed gasdynamics [3], atmospheric physics [4], acoustics [5], chemodynamics [6~8] and so forth. In the past ten years, a series of research concerning the vibrational relaxation process was widely conducted. A series of papers [9 ~ 13] as well as review articles [1, 14 ~ 17] were presented. The relaxation process has created a key effect on gas laser application. Without sufficient

knowledge about this relaxation process, it is impossible to invent a new laser system, to improve device performance or to conduct analysis of the laser dynamic process. Laser technology development requires in-depth understanding about the relaxation process; however, it itself has become a powerful measuring tool for the relaxation process experiment.

At the beginning of 1969, a review concerning the vibrational relaxation process of a $\text{CO}_2\text{-N}_2$ laser system was presented by Taylor and Bitterman [1]. This review is still widely recommended today; however, it lacks data for many important relaxation processes because of the research level at that time. Data process and analysis were imperfect which made some of the rate data inaccurate; moreover, some important problems in the relaxation process of the system were not thoroughly studied. Since 1969, a wealth of data were accumulated for the relaxation process of the $\text{CO}_2\text{-N}_2\text{-H}_2\text{O}$ laser system. In order to calculate the performance of a gas dynamics laser (GDL), some new data were collected and used [18,19]; however, no detailed analysis for the relaxation process was performed. The quantity of collected data was also little; only some very imperfect rate values were presented, and thus the calculation results of the GDL performance showed large errors [2].

Based on Reference [1], all new experiment data and analytical results from then until the present are further collected and processed; the best curve fits and analytical formula for various



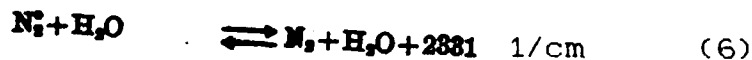
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relaxation rates are presented, and thus can be easily to used. The best curve fits presented in this paper extend the data applicable range, and point out the major difference from the Ref. [1] results. Good results are also obtained by applying this paper's results to the non-equilibrium calculation of a burning type GDL. This means that the reliability of this paper's results is again proved from an additional aspect.

II. Relaxation Mechanism

The energy exchanging reactions considered here are (see Fig. 1):

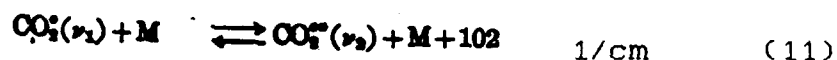
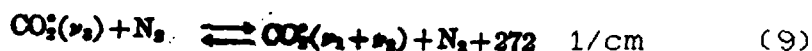
V-T exchange:



Intermolecule V-V exchange:



V-V exchange inside molecule:



where * represents the vibrational energy particle; the symbol inside the brackets indicates the CO vibration mode. The energy difference is taken from translation or rotation.

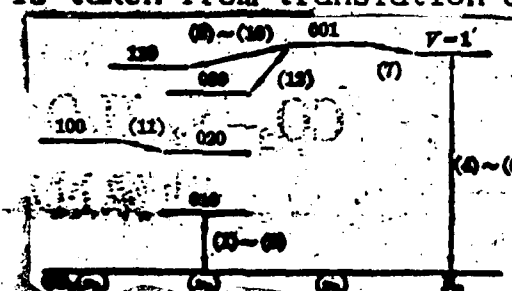


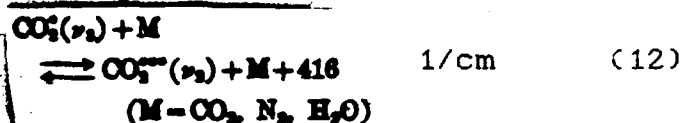
Fig. 1 Illustration of the energy level of a CO -N laser system. 2 2

Only a few of the lower vibrational energy levels of the system are presented in the formulae and figures. If the relaxation equation of vibrational energy is used to describe the system, the effects of all similar processes can be included [20]; if an equation which considers the particle number only at the lowest energy level is used to describe the relaxation process,

errors can occur. However, for the temperature range ($<1500\text{K}$) in which we are interested, this approximation is acceptable.

The relaxation mechanism of a molecule system is extremely complicated. Herzfeld [11] has analytically calculated in detail for the CO_2 system based on the SSH theory [5] and gives the jumping probability for all possible processes. However, the experiment results indicate that the SSH theory has a lot of constraints. The fact is that the jumping probability among various vibrational states is difficult to measure directly, and many people are currently devoting themselves to determining the relaxation mechanism indirectly by using a laser method.

The $\nu_1 \rightarrow \nu_1 + \nu_2$ mechanism is adopted to study the V-V exchange inside a $\text{CO}_2(\nu_1 \rightarrow \nu_2)$ molecule while the $\nu_1 \rightarrow 3\nu_2$ mechanism is used in Ref. [1].



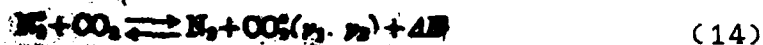
Under an equilibrium condition between $\text{CO}_2(\nu_1)$ and (ν_2) modes, it can be proved that there exists a simple relation between the rates in Equation (8) to (10) and (12) [22],

$$\begin{aligned} & K[\text{O}_3(1 \rightarrow 0), \text{O}_2(0 \rightarrow 3), \text{M}] \\ &= \frac{(1 - e^{-\theta_1/T})^2}{1 - e^{-\theta_2/T}} \\ & \times K[\text{O}_3(1 \rightarrow 0), \text{O}_1(0 \rightarrow 1), \\ & \quad \text{O}_2(0 \rightarrow 1), \text{M}] \end{aligned} \quad (13)$$

where θ_i is a vibrational characteristic temperature. Thus only

the reaction rates of Equation (8) to (10) are given here.

From the experiment data analysis, it shows that the rate of



is zero in the error range [23], thus it is ignored.

A small amount of water has a great effect on the relaxation process of a CO_2-N_2 system. Due to the difficulties of experiments, imperfect theory and the complications of water reaction, the relaxation mechanism after adding water is still not clear. In summary, there are two viewpoints: one states that a V-V exchange first occurs between CO_2^* and N_2^* , and then the H_2O performs V-T excitation (see Ref. [1]); the other one considers the water molecule as a collision companion in the V-T or V-V excitation of CO_2^* , N_2^* . The latter theory is adopted in this paper. If the V-T excitation rate for H_2O^* is extremely great, it can be proved that the water vibration mode can be treated as in equilibrium with the translation, thus the effects from both viewpoints are equivalent.

III Experiment and Data Process

There are many experimental methods for studying vibrational relaxation. The most common methods are a shock tube (compressible flow), laser fluorescence, laser photosound, ultrasound, expansion flow in a shock tube, collision tube,

etc..

The relaxation rate is a microscopic datum which describes one of the characteristics of a molecular vibrational energy level and cannot be measured directly from experiments. The measurement in an experiment is some sort of macroscopic physical quantity which has a close relation to the relaxation process of interior degree-of-freedom. A certain theoretical analysis is necessary to connect the experiment data (such as decay constant or total relaxation time) with the relaxation rate of some processes. Obviously, the speed accuracy also depends on data process in addition to the experiment itself.

The Schafer formula for the V-T rate of a multi-atom molecule [24] is always used,

$$\tau_1 = \tau_2 \times \frac{C_1}{\sum_i C_i} \quad (15)$$

where the subscript 1 represents the mode of "control relaxation rate"; C_1 is the vibrational specific heat. Study [22] shows that this formula is good only when the V-V rate in the system is much greater than the V-T rate, a "control rate"; however, the actual V-V rate is not sufficiently large. The error is certainly introduced when the Schafer formula is adopted, especially under the condition of small specific heat. This error, however, is small in a CO₂ system.

The V-V rate process was recently developed. Under the condition of a pure CO and CO-H₂O mixture, the decay constants, k , which is measured by a shock tube experiment [25] performed by Taylor and Bitterman, and the laser fluorescence experiment carried by Rosser et. al. [23, 26] are then converted to rate coefficient through the formula,

$$K[CO(1 \rightarrow 0), CO(0 \rightarrow 3); M] = \frac{k}{2b} \quad (16)$$

Based on the common form of the relaxation equations [20], a relaxation mechanism is analyzed. A solution is also obtained by solving a set of relaxation equations [22],

$$K = \frac{kK[CO(1 \rightarrow 0), O] - k'(1+o)}{K[CO(1 \rightarrow 0), O] - k(a+b+o)} \quad (17)$$

where

$$a = \frac{1}{2(\rho^2 - 1)(\rho^2 - 1)} \quad (18)$$

$$b = \frac{1 - \rho^2}{(1 - \rho^2)^2} \quad (19)$$

$$c = \frac{1 - \rho^2}{(1 - \rho^2)^2} \quad (20)$$

According to Ref. [20], the factor 2 in the Taylor and Bitterman formula is combined into the constant K . If the $K[CO(1 \rightarrow 0)]$ in Equation (17) is much greater than k , it can be reduced to Equation (16). This is the exact approximation adopted by Taylor,

Bitterman and Rosser which considers that $\text{CO}(\nu_2)_T$ and translation are in balance. The rate obtained by using Equation (16) will have an error in the relation of its numerical value and the temperature. It is more complicated for the $\text{CO}_2\text{-N}_2$ mixture, both the rate of $\text{CO}_2(\nu_2)_T$ and the rate of $\text{CO}_2(\nu_2)_T \rightarrow \text{N}_2$ have to be solve simultaneously. A similar approximation was adopted by them, and thus errors were created. The results processed by these two formulae are presented in this paper.

The unit of rate K used in this paper is $\text{cm}^3/\text{molecule}\cdot\text{sec}$. The relation between K and the jumping probability P, the formula which converts various relaxation experiment data to K, and those necessary physical constants are listed in Reference [1].

IV Relaxation Speed Data

Since the vibrational relaxation time is short, and the requirements of the experiment (such as impurity content) are strict, the data measured from experiments often show large errors; some time these errors are one or two orders of magnitude. According to the data reliability and experiment accuracy analyses, a certain weight is added to various data measured by different authors, and then a weighted least square method is used to get the best fit in the form of

$$\log_{10} K = A + BX + CX^2 + DX^3 \quad (21)$$

(all calculations are performed in a state-made computer TQ-16).
According to Landau-Teller and SSH theory, generally, the X is
chosen as $T^{-1/3}$. Under a high temperature condition, should
be linear to $T^{-1/3}$.

Table 1 Best fitting coefficients

(1)	(2)						(3)
过 程	速率	X	A	B	C	D	温度范围
(1) $\text{CO}_2(\nu_1 \rightarrow T) - \text{CO}_2$	K	$T^{-1/3}$	-7.22788741	-77.0945460	203.852916	0	150~7000
(2) $\text{CO}_2(\nu_1 \rightarrow T) - \text{N}_2$	K	$T^{-1/3}$	-9.18704456	-42.4606794	46.4011141	0	150~1500
(3) $\text{CO}_2(\nu_1 \rightarrow T) - \text{H}_2\text{O}$	K	$T^{-1/3}$	-9.827809	-25.22583	125.6784	0	300~1000
(4) $\text{N}_2(\nu \rightarrow T) - \text{N}_2$	K	$T^{-1/3}$	-5.38873	-139.0598	88.50646	1412.096	240~4700
(5) $\text{N}_2(\nu \rightarrow T) - \text{CO}_2$	K	$T^{-1/3}$	-5.38873	-139.0598	88.50646	1412.096	240~4700
(6) $\text{N}_2(\nu \rightarrow T) - \text{H}_2\text{O}$	K	$T^{-1/3}$	-10.9754595	-21.7181799	0	0	290~3000
(7) $\text{CO}_2(\nu_2) \rightarrow \text{N}_2$	K	$10^{-3} \cdot T$	-12.0624762	-0.0980571825	5.539994×10^{-5}	-6.277832×10^{-5}	150~2600
(8) $\text{CO}_2(\nu_1 \rightarrow \nu_1 + \nu_2) - \text{CO}_2$	K_{TS}	$T^{-1/3}$	-9.25923236	-4.69827095	-470.105702	1930.79807	150~1600
	K	$T^{-1/3}$	-9.7767526	2.92817157	-504.993077	1978.16692	150~1600
(9) $\text{CO}_2(\nu_1 \rightarrow \nu_1 + \nu_2) - \text{N}_2$	K_{TS}	$T^{-1/3}$	5.61975614	-347.252296	2057.64553	-4233.96898	160~1600
	K	$T^{-1/3}$	7.42712139	-390.990028	2387.44778	-5029.38366	160~1600
(10) $\text{CO}_2(\nu_1 \rightarrow \nu_1 + \nu_2) - \text{H}_2\text{O}$	K_{TS}	$T^{-1/3}$	-14.1877762	38.1508547	-159.940244	0	300~1000
	K	$T^{-1/3}$	-13.8868956	38.8333884	-144.499031	0	300~1000
(11) $\text{CO}_2(\nu_1 \rightarrow 2\nu_2) - \text{CO}_2$	(4)慢K	T	-13.07233	0.00137833	0	0	300~1000
	(5)快K	认为 $K=1.25 \times 10^{-11}$, 且不随温度改变 (6)					
(12) $\text{CO}_2(\nu_1 \rightarrow 2\nu_2) - \text{N}_2$	一种认为与 $\text{CO}_2(\nu_1 \rightarrow 2\nu_2) - \text{CO}_2$ 的速率近似相等 (7)						400
	另一种认为是 $\text{CO}_2(\nu_1 \rightarrow 2\nu_2) - \text{CO}_2$ 的速率的 0.46 倍 (8)						400
(13) $\text{CO}_2(\nu_1 \rightarrow 2\nu_2) - \text{H}_2\text{O}$	缺乏数据 (9)						

Key: (1) Process; (2) Rate; (3) Temperature range;
(4) Slow K; (5) Fast K; (6) Considered as
 $K=1.25 \times 10^{-11}$, and not varied with temperature; (7)
Considered as equal to the $\text{CO}_2(\nu_1 \rightarrow 2\nu_2) - \text{CO}_2$ rate; (8)
Considered as 0.46 time of $\text{CO}_2(\nu_1 \rightarrow 2\nu_2) - \text{CO}_2$ rate; (9)
Lack of data.

The best fit coefficient is list in Table 1. Figure 2 to 10 are plots of the relation between rate coefficients and temperatures.

Anderson [18] investigated the effects of various relaxation rate uncertainties on the GDL performance by using his simplified relaxation model and the base of the linear relaxation equation. He concluded that the reactions of Equation (3), (10), (6) and (9) have the most effect.

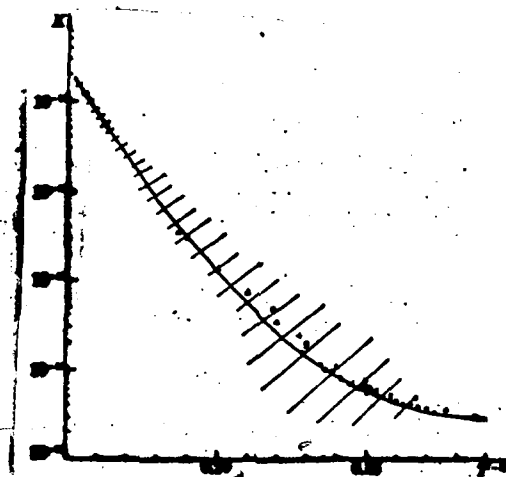
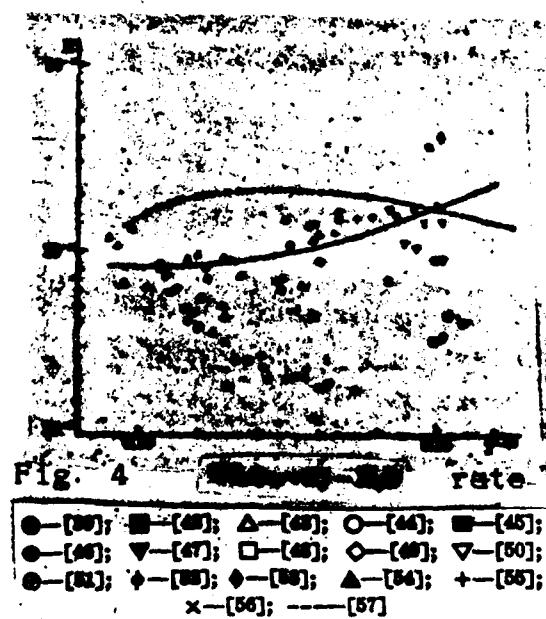
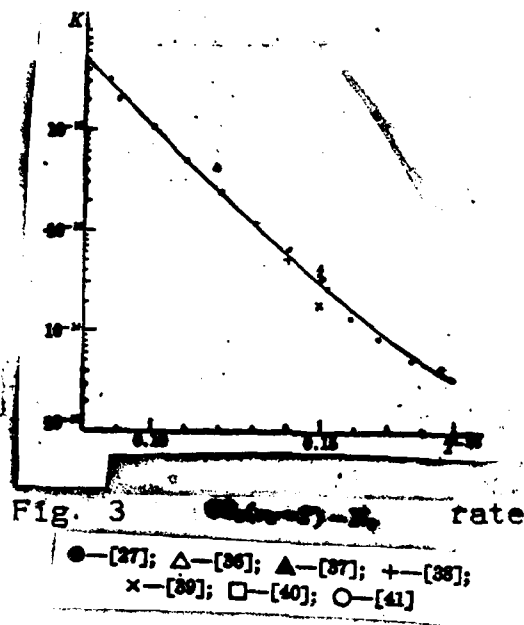


Fig. 2 $\text{CO}_2(\text{m-F}) - \text{CO}_2$ rate

$\square$ —[1]; Δ —[27]; $+$ —[28]; $\blacksquare$ —[29];
 $\square$ —[30]; Δ —[31]; $\circ$ —[32]; $\bullet$ —[33]
 $\times$ —[34]



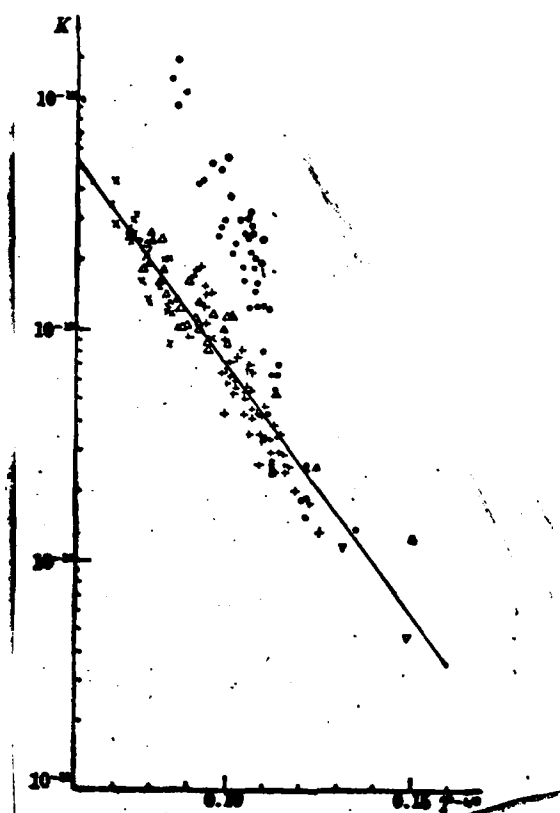


Fig. 5 rate

●—[28]; ○—[30]; △—[32]; ▽—[31];
x—[33]; ▽—[35]; ▲—[34]; +—[36]

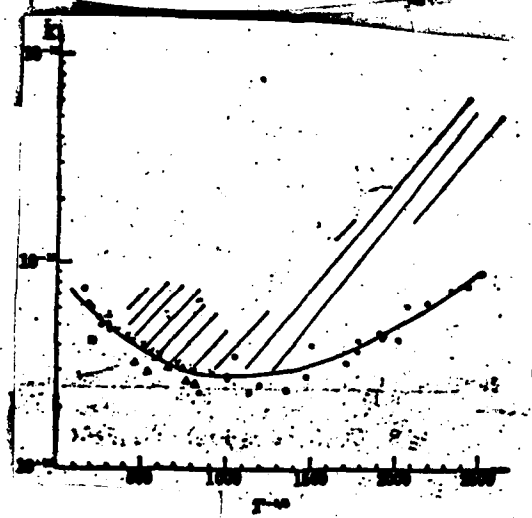


Fig. 6 $\text{CO}_2(p_0) \rightarrow \text{H}_2$ rate

x—[28]; ///—[25]; △—[44]; +—[37];
▲—[68]; ○—[69]; ●—[70]; □—[71]

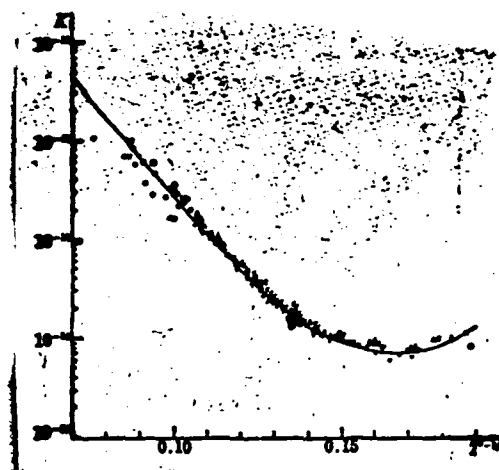


Fig. 7 $\text{CO}_2(\nu_2 \rightarrow \nu_1 + \nu_2) - \text{CO}_2$ rate

○—[23]; ◆—[68]; ■—[70]; ▼—[71]; ⊗—[72];
 +—[73]; ●—[74]; ⊙—[75]; □—[76]; ×—[77];
 △—[78]; ◆—[79]; ▲—[80]; ▽—[81]; ◇—[82]

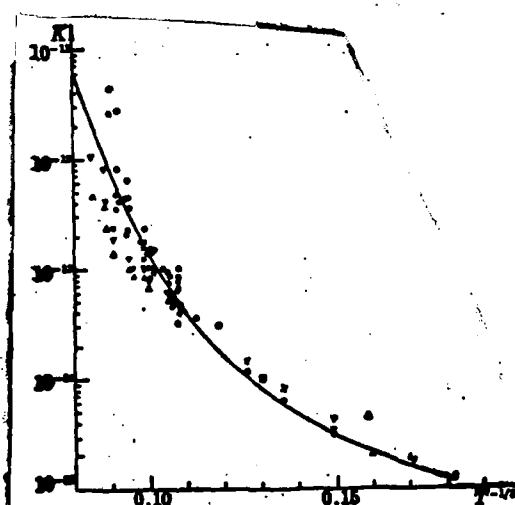
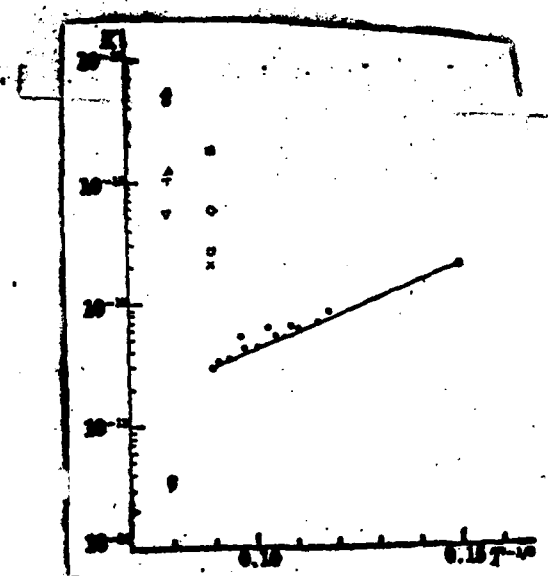
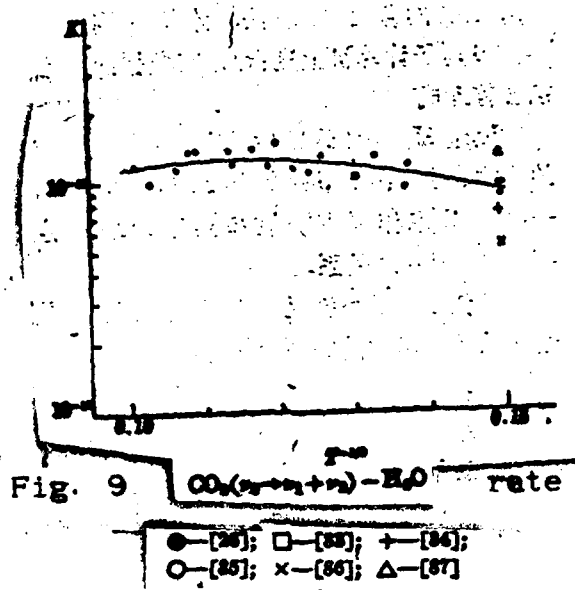


Fig. 8 $\text{CO}_2(\nu_2 \rightarrow \nu_1 + \nu_2) - \text{N}_2$ rate

○—[23]; ●—[25]; +—[40]; △—[69, 8-2];
 ▽—[69, 8-3]; ▲—[70]; ■—[71]; ×—[72];
 ▼—[77]; □—[83]



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