



## Plasmadynamics and Lasers Award Lecture

The Ohio State University

Nonequilibrium Thermodynamics Laboratories

# Thermal Mode Nonequilibrium in Gas Dynamic and Plasma Flows

by

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The Ohio State University*

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Seattle, Washington, June 25, 2008

- **What is thermal mode nonequilibrium?**  
**Emphasis on high density, collision-dominated flows,  
cold molecular plasmas**
- **Environments**
  - i) Cool Electric Discharge Plasmas**  
**Self-Sustained vs. Preionized**
  - ii) Optically Pumped Plasmas**  
**Excitation of Vibrational States**  
**Kinetics and Energy Transfer Studies**
  - iii) Gas Dynamic Flows**  
**Supersonic Expansions of High Enthalpy Gases**  
**Strong Shock Waves**
- **Applications**
  - i) Plasma Wind Tunnels**
  - ii) Gas Lasers**
  - iii) Chemistry: Isotope Separation**
- **Summary: Where do we go with this?**

# What is thermal mode equilibrium?

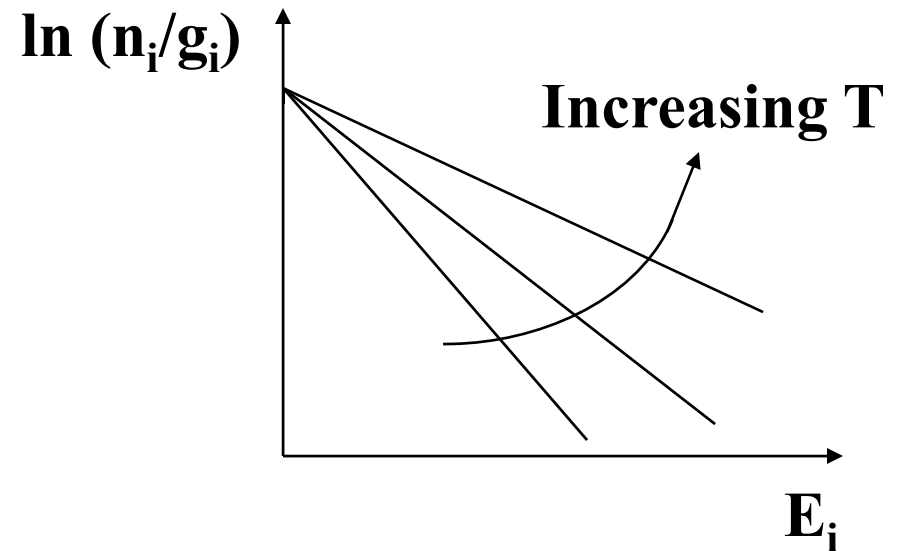
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$$n_i/g_i \sim \exp ( - E_i/kT ),$$

$$N = \sum n_i , \text{ and } E = \sum n_i E_i$$

If we know the energy levels,  $E_i$ ,  
and the gas temperature,  $T$ , we can  
calculate the whole ideal gas  
thermodynamic table:



**Table 1 Air at Low Pressures (for One Pound)**

<b>T</b>	<b>t</b>	<b>h</b>	<b>p<sub>r</sub></b>	<b>u</b>	<b>v<sub>r</sub></b>	<b>φ</b>
<b>1000</b>	540.3	240.98	12.298	172.43	30.12	.75042
<b>1001</b>		241.23	12.343	172.61	30.04	.75067
<b>1002</b>		241.48	12.388	172.79	29.96	.75092
<b>1003</b>		241.73	12.433	172.97	29.88	.75117
<b>1004</b>		241.98	12.478	173.15	29.81	.75142

# What is thermal mode nonequilibrium?

**Thermal nonequilibrium exists whenever:**

- The populations,  $n_i/N$ , of one or more modes are distributed according to the Boltzmann law, *but* the temperature,  $T$ , of at least one mode differs from that of the others.

**OR**

- The populations,  $n_i/N$ , of one or more modes are *not* distributed according to the Boltzmann law.

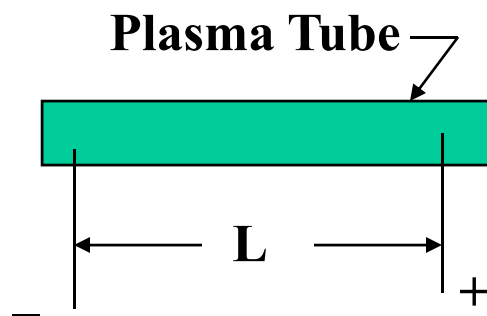
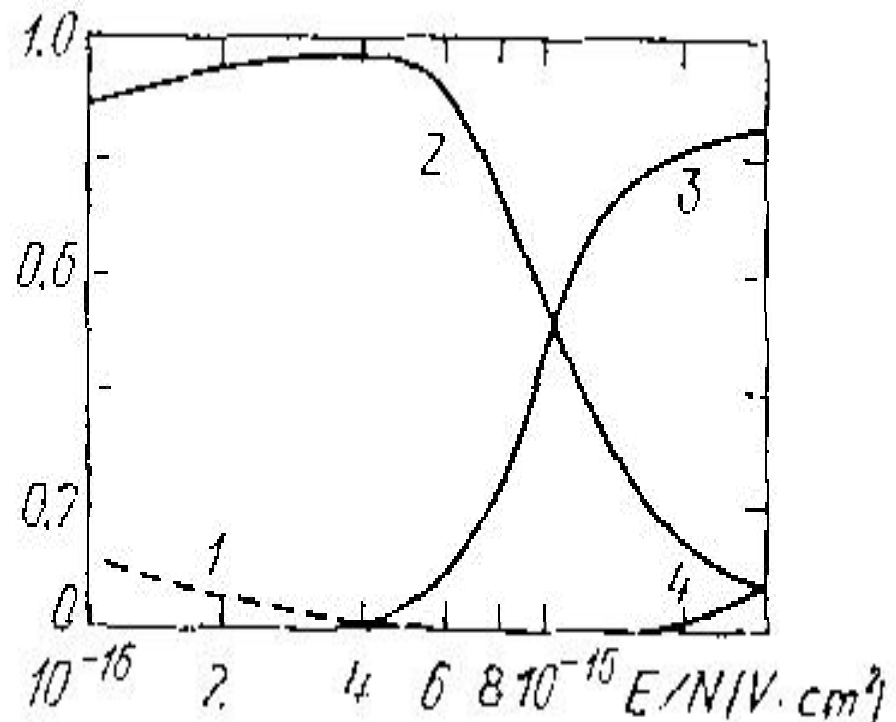
# Environments: Energy Input Among Nonequilibrium Plasma Modes in a Self-Sustained Electrical Glow Discharge

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**Fraction of Power into each Mode, as a function of  $E/N$ .  $E/N$  is approximately proportional to the mean energy of the free electrons.**

1. Rotational mode
2. Vibrational mode
3. Electronic mode
4. Ionization



**Ratio of Electric Field,  $E$ , to total number density of molecules,  $N$**

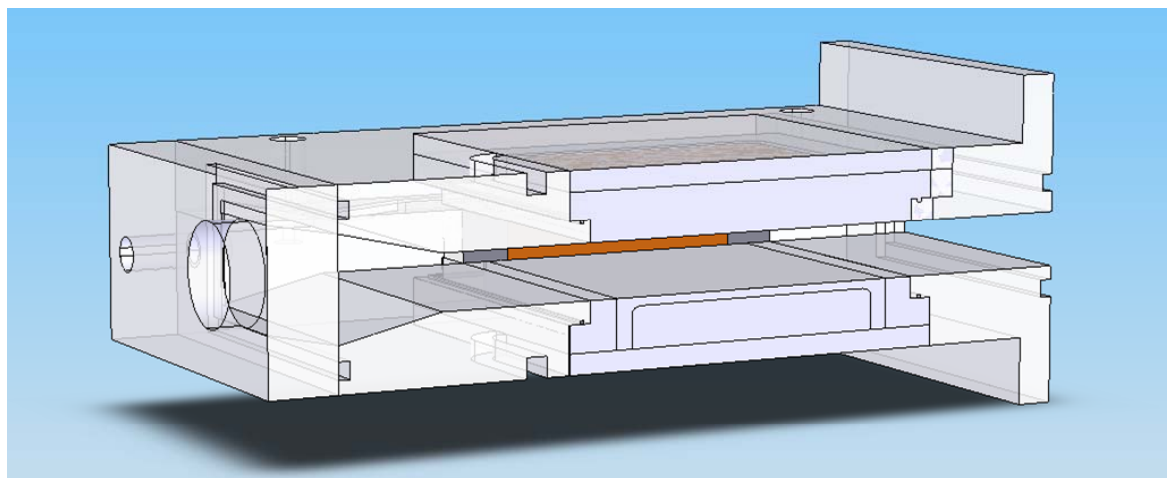
**Electric Field = Voltage/Electrode Separation**  

$$E = V/L$$

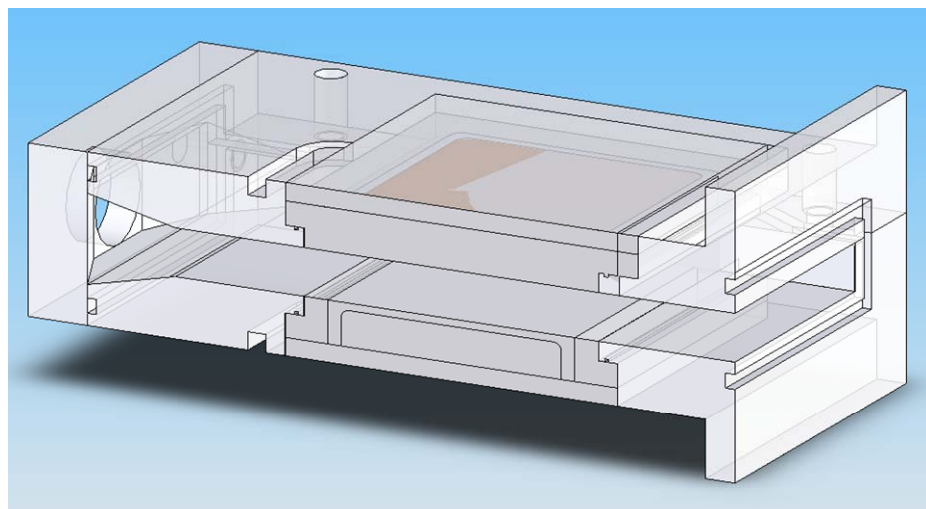
## **Environments: Preionized Electrical Glow Discharge** **Short Pulse HV Ionizer, D.C. Sustainer - Geometry**

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**Pulser electrodes  
in top and bottom walls  
DC electrodes in side walls  
Flow direction left to right**



**Dimensions: 1 cm x 5 cm, 10 cm long**

**Mach number:  $M \sim 0.2$**

**Discharge pressure: up to 160 torr**

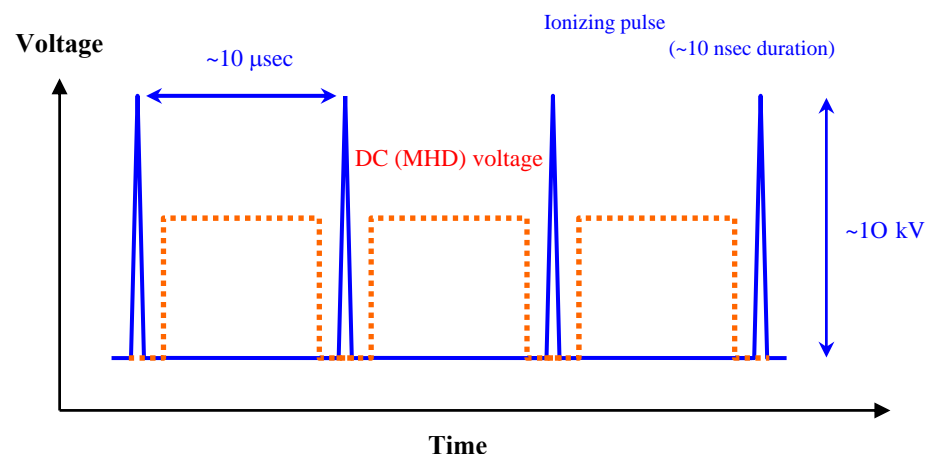
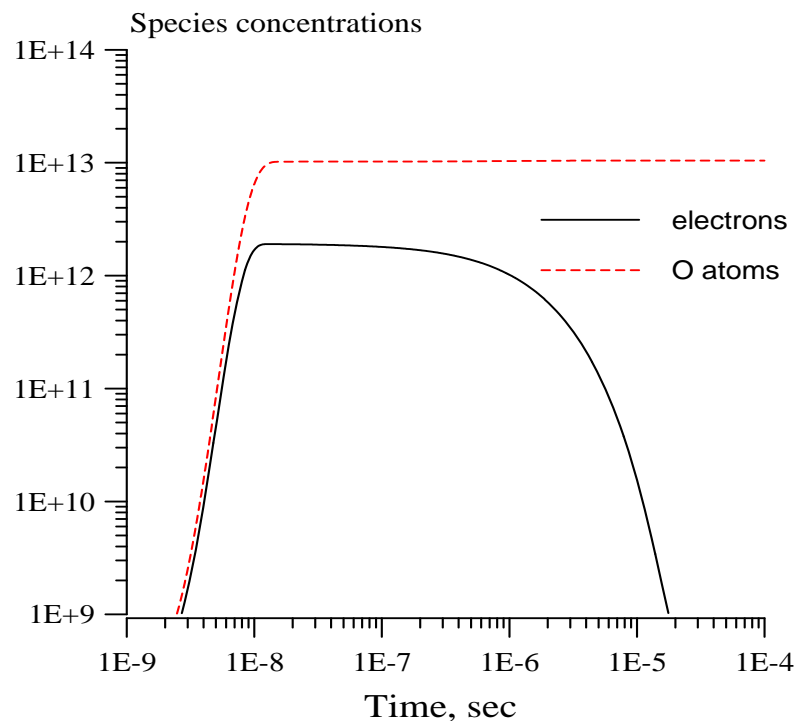
**( $O_2$ -He,  $O_2$ -Ar)**

**Mass flow rate: up to 12 g/sec**

# Environments: Preionized Electrical Glow Discharge Short Pulse HV Ionizer, D.C. Sustainer -Performance

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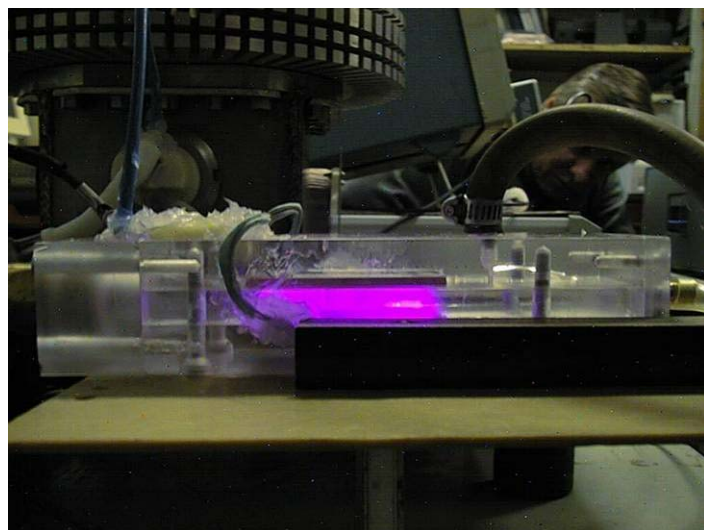
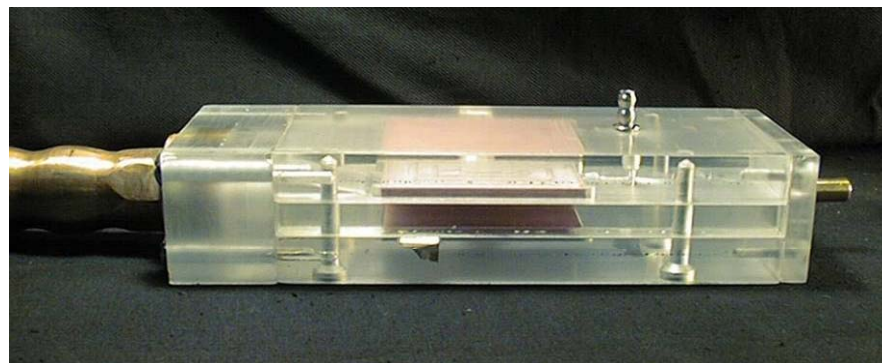
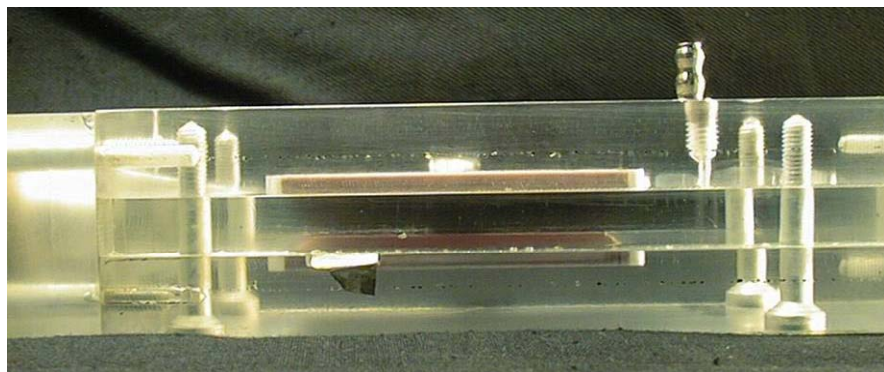
**On the left, species concentrations after the ionizing pulse, on the right, schematic of the pulsed and d.c. discharge operation.  $P=0.1$  atm,  $T=300$  K,  $E/N_{\max}=350$  Td,  $E_{\max}=8.5$  kV/cm,  $\tau_{\text{pulse}}=10$  nsec**



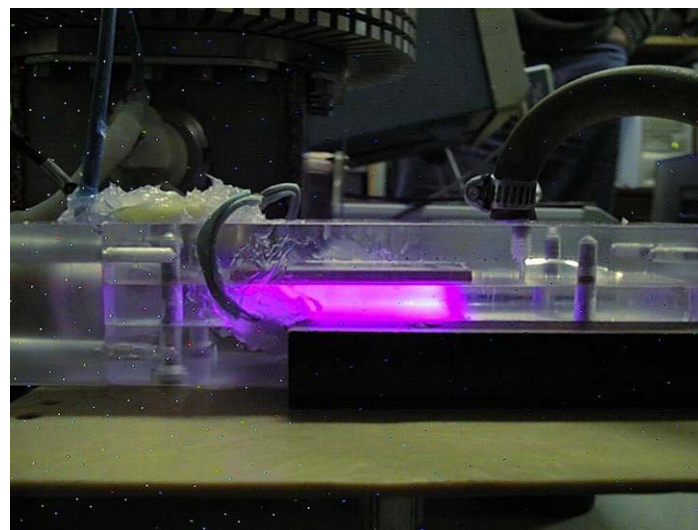
## **Environments: Preionized Electrical Glow Discharge** **Short Pulse HV Ionizer, D.C. Sustainer -Photos**

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**P=300 torr, 10 kHz**



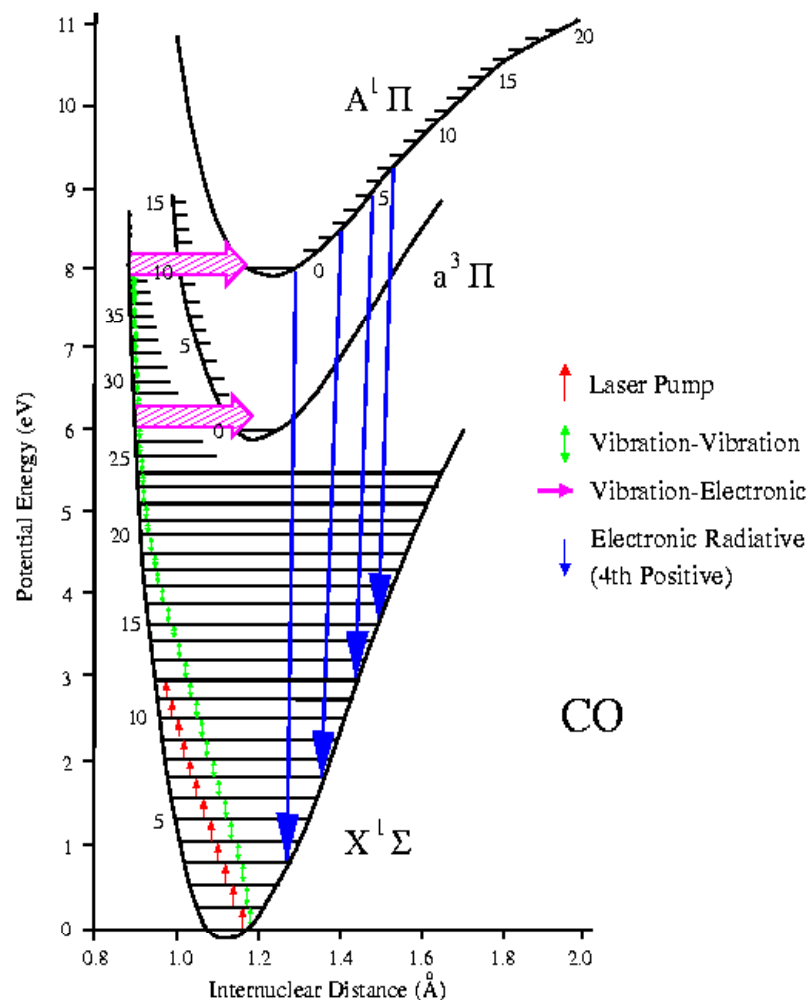
**P=500 torr, 10 kHz**



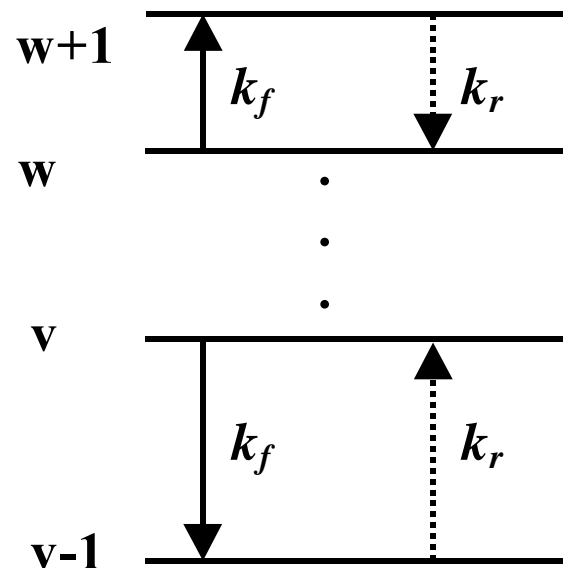
# Environments: Optical Pumping – Excitation of Vibrational Mode by CO Laser Radiation. Vibration-Vibration (V-V) Exchange

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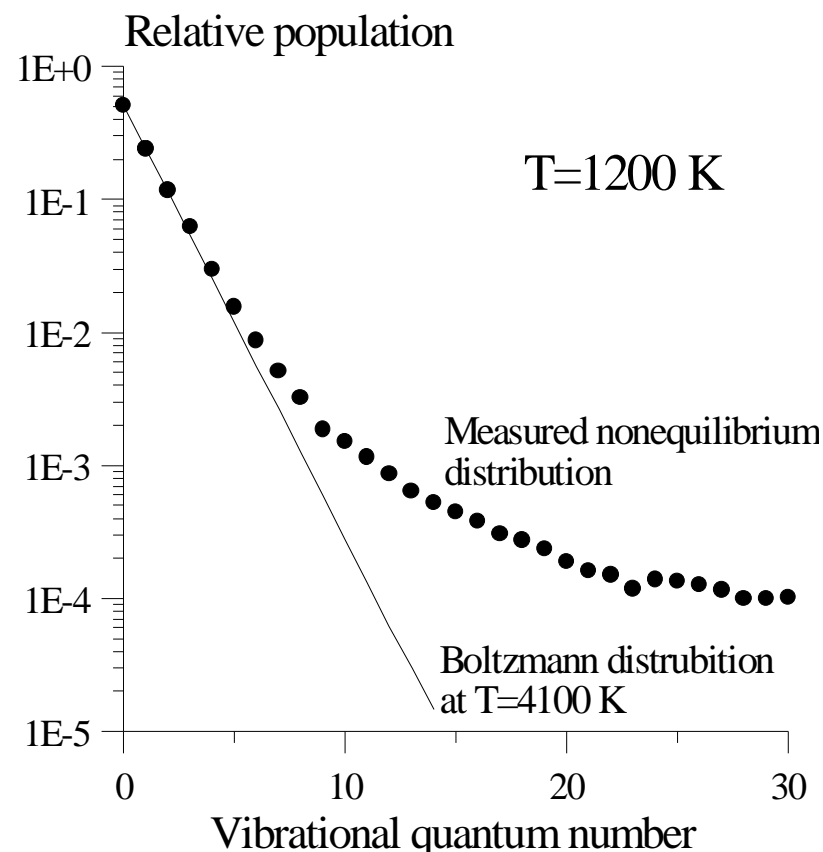
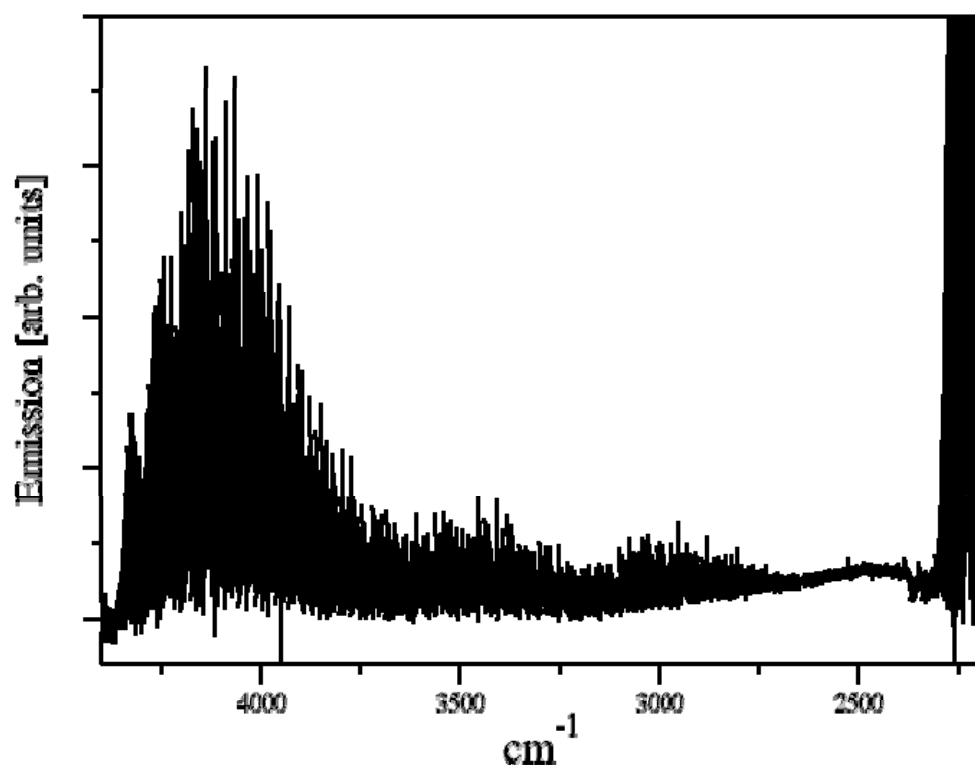
$$\frac{k_k}{k_r} = \exp\left(\frac{\Delta E_{v,v-1} - \Delta E_{w+1,w}}{T}\right) > 1$$



# Environments: Optical Pumping – Excitation of Vibrational Mode by CO Laser Radiation. Measurement of Vibrational State Populations – Vibrational Distribution Function

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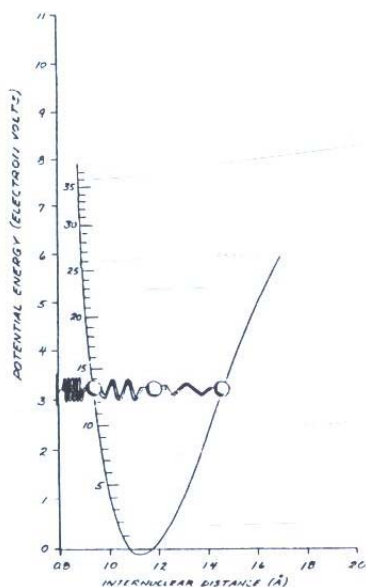
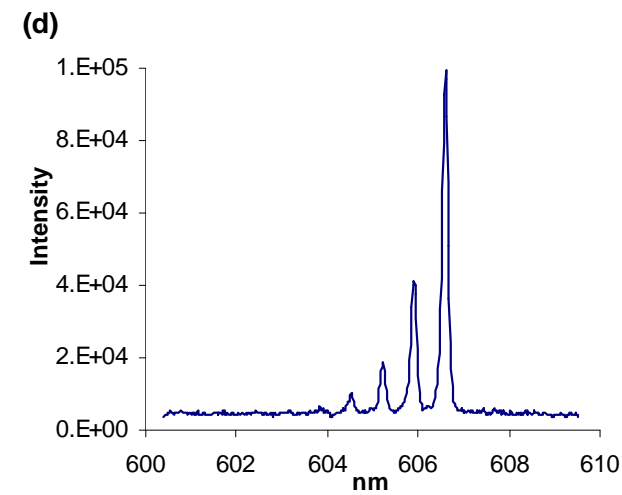
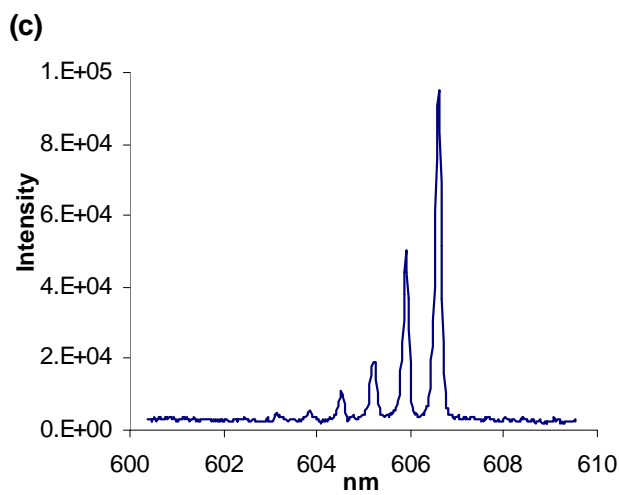
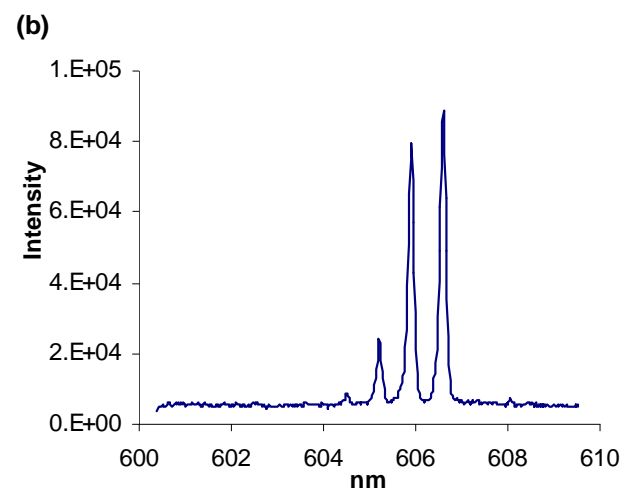
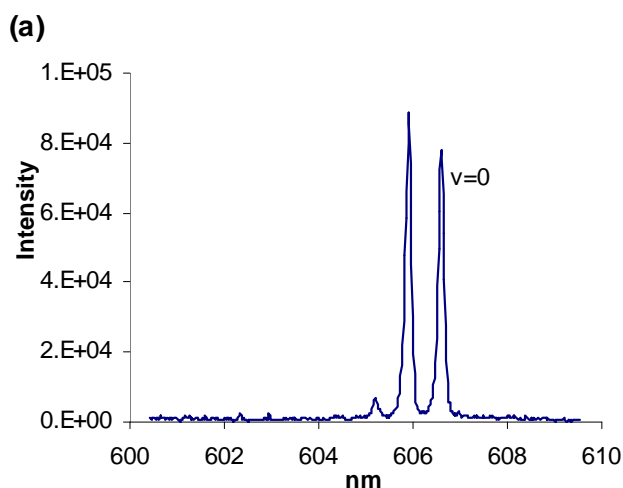


# Environments: Optical Pumping of Nitrogen Vibration by Raman Absorption – Excitation of High Vibrational States by V-V Energy Exchange

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Each peak is from a successive vibrational state,  $v$ , ground state signal on the right. The time delay between the pump and probe pulse is (a) 150 ns, (b) 1 ms, (c) 5 ms, (d) 10 ms.

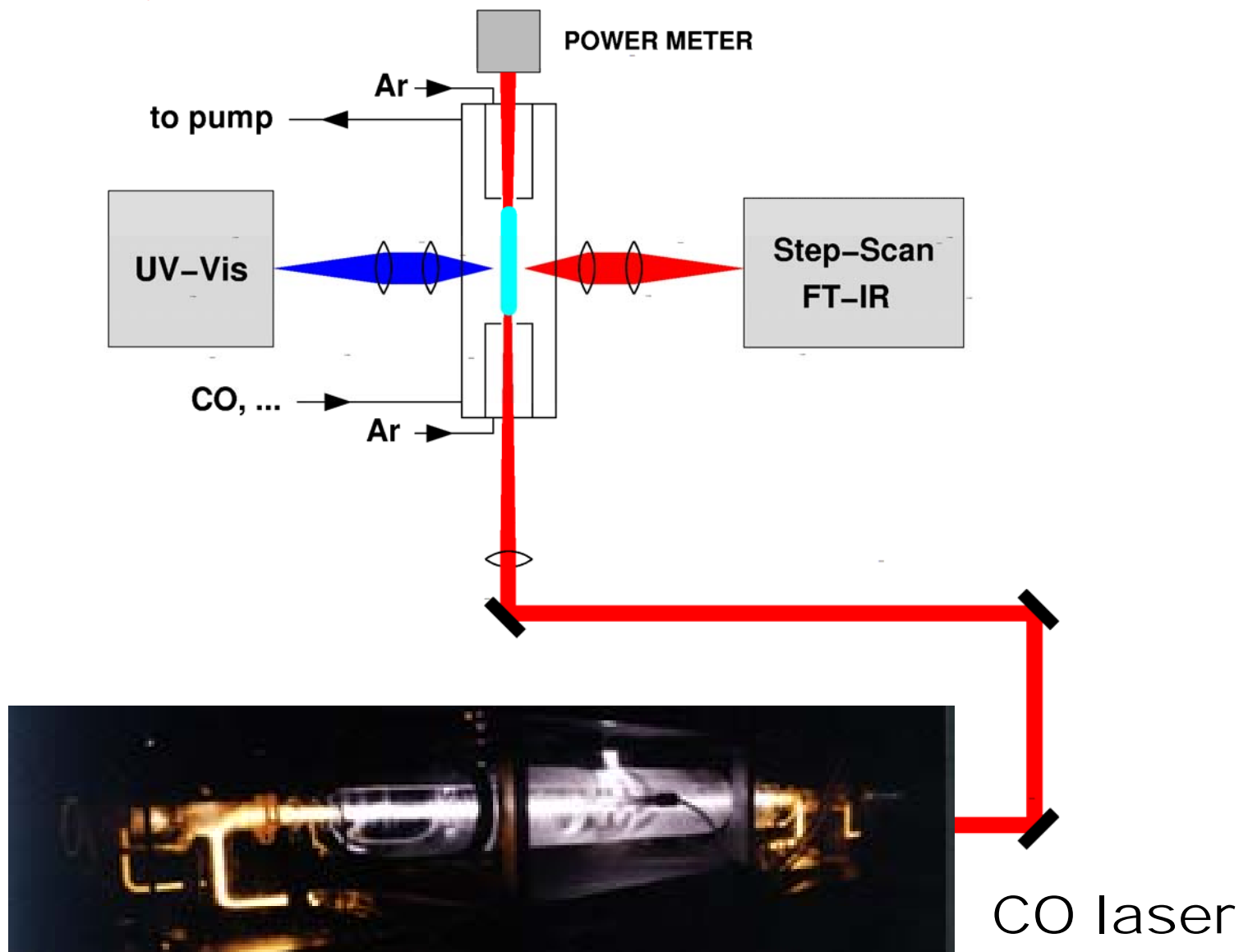


# Optically Pumped Plasmas: Kinetics Studies

## Experimental Setup for CO Plasma

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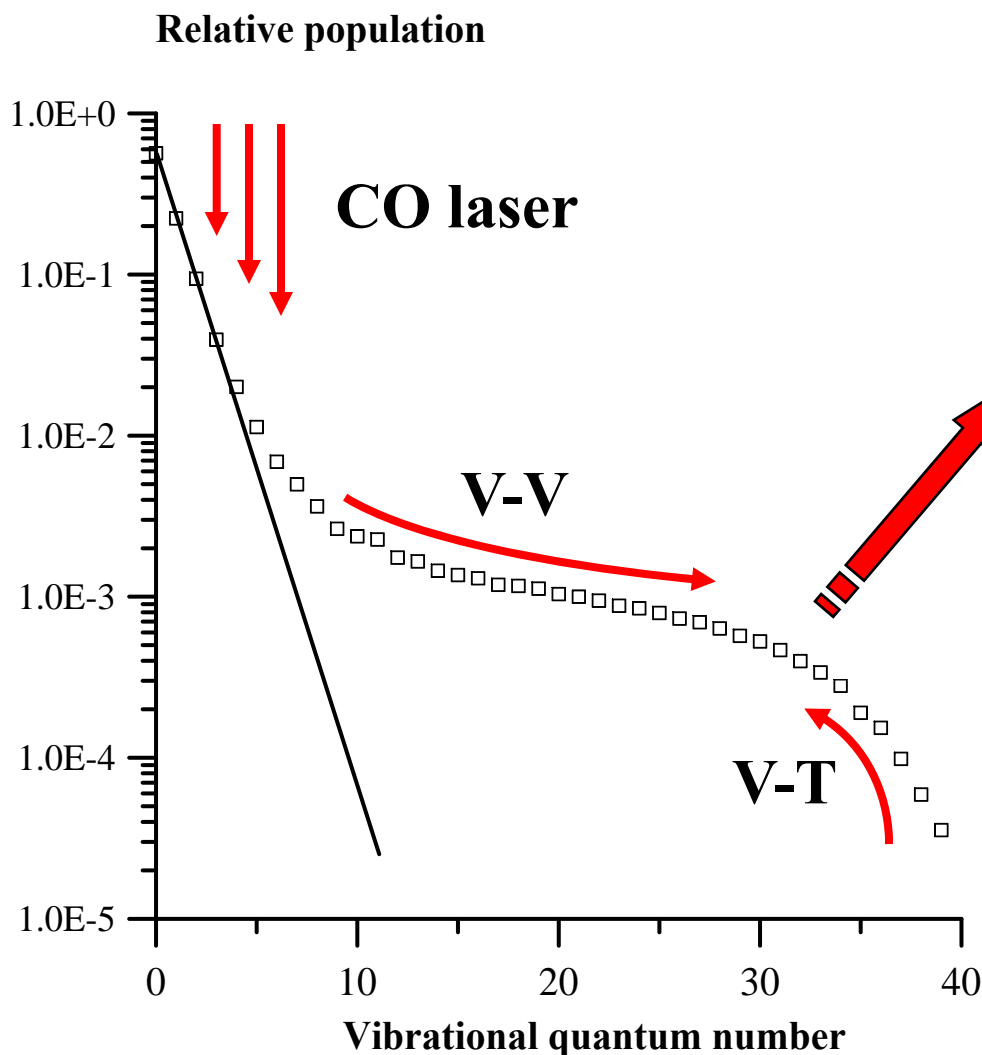


# Optically Pumped Plasmas: Kinetics Studies

## Energy Transfer and Kinetics Processes in a CO Plasma

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2 Torr CO, 100 Torr Ar,  
CO laser power 10 W c.w.

**T = 600 K**  
**T<sub>v</sub> = 3300 K**

**Ionization:**



**V-E:**



**Chem. Reactions:**

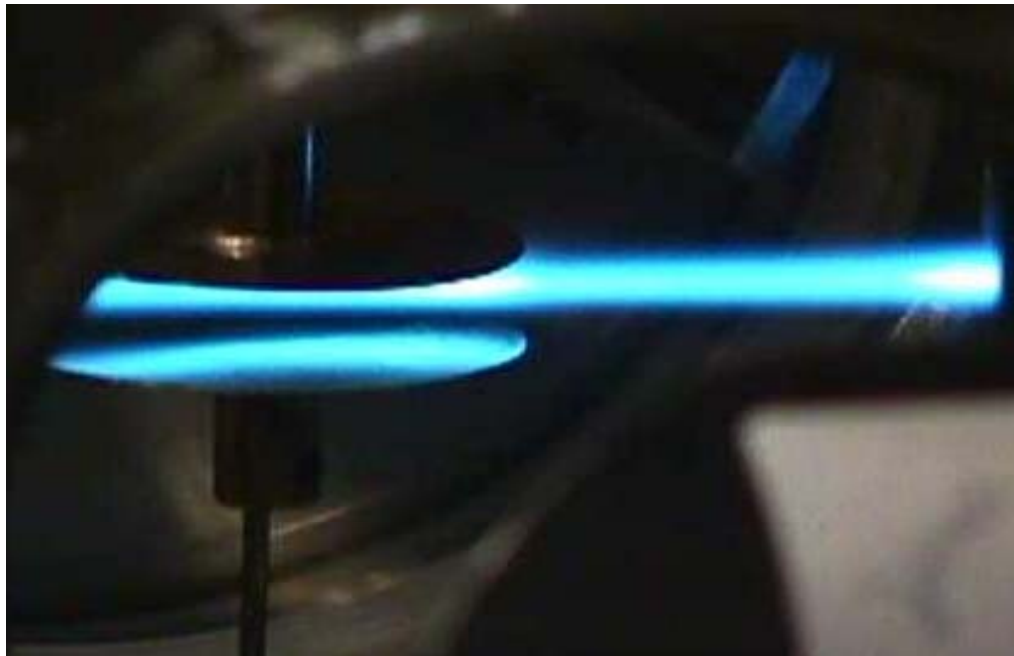


# Optically Pumped Plasmas: Kinetics Studies

## Optically Pumped CO Plasma Photo

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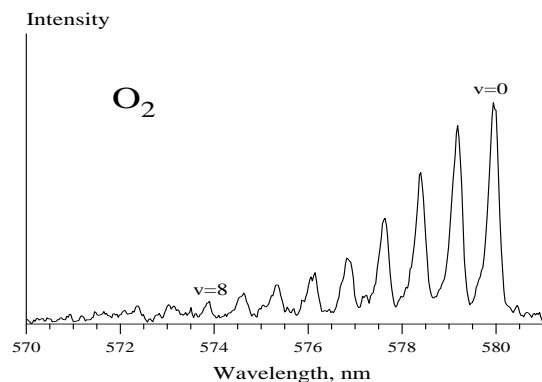
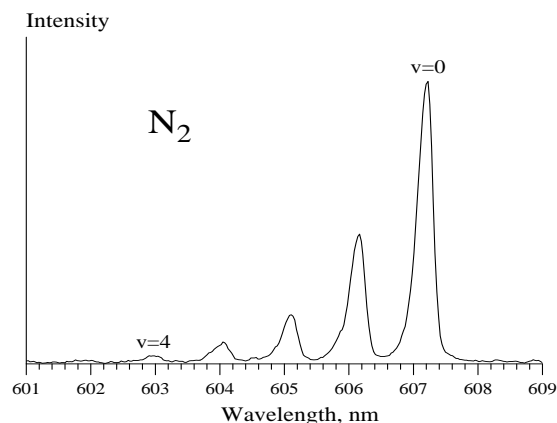
# Optically Pumped Plasmas: Kinetics Studies

## Steady –State Vibrational Excitation of a Flowing Dry Air Mixture

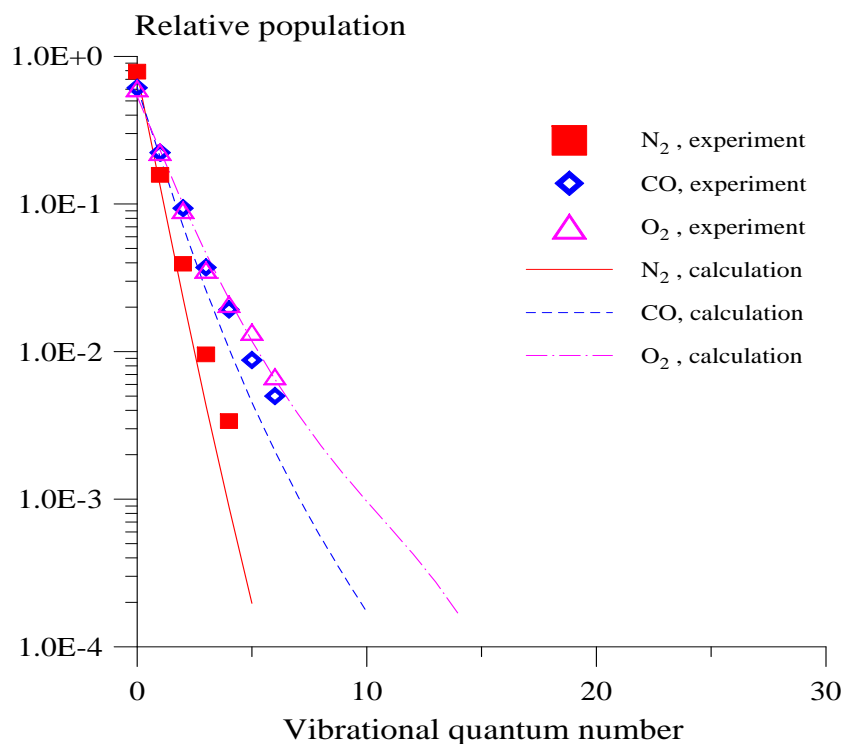
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Vibrational energy level populations for an optically pumped air plasma. If energy were equilibrated, temperature of vibrational modes would be  $\sim 2000\text{K}$ . Measured as kinetic temperature is only  $500\text{ K}$ . Extreme nonequilibrium is created in a steady state, atmospheric pressure, molecular gas plasma. Gas mixture is  $\text{CO}/\text{N}_2/\text{O}_2=5/75/20$ ,  $P=1\text{ atm}$ . Pump laser power is  $10\text{ W}$ .

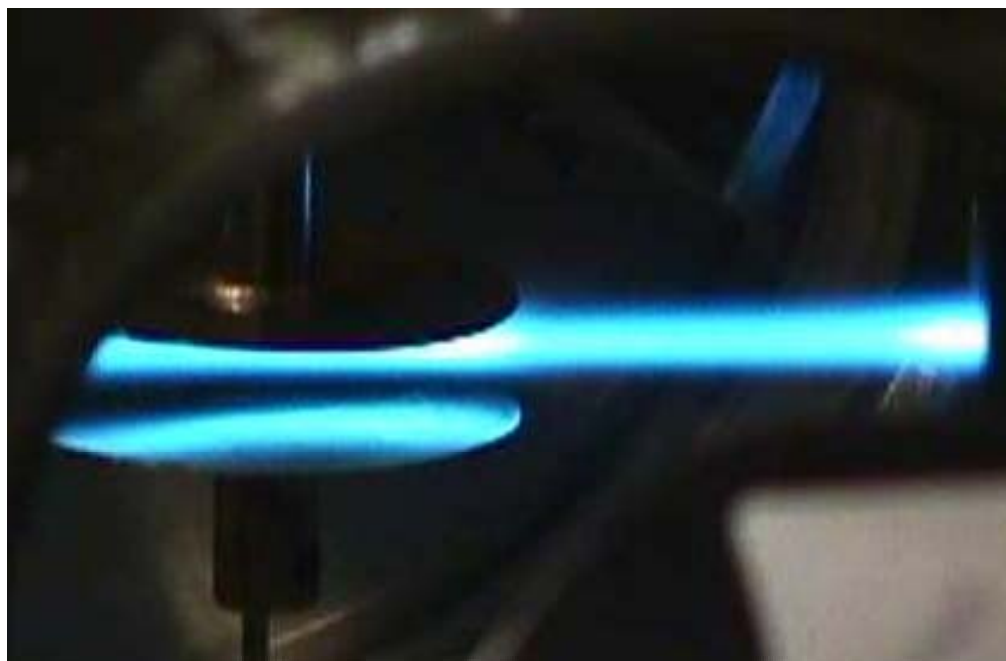


Raman Spectra



Experimental Vibrational State Populations  
Inferred from Raman Spectra. Calculated State  
Populations from Master Equation Kinetic Model

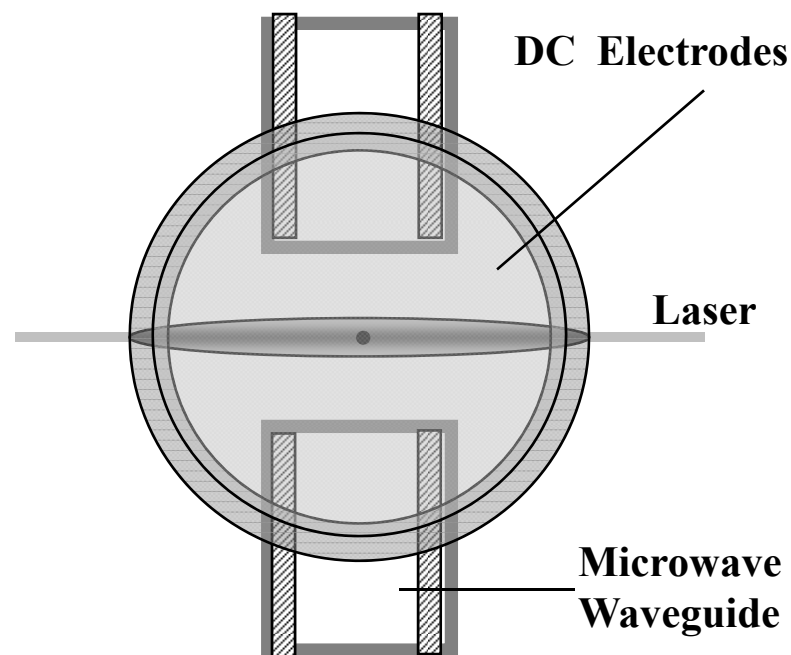
### Thomson discharge



**Electron production rate:**

$$k_{\text{ion}} \sim 10^{-13} \text{ cm}^3/\text{s}$$

### Microwave absorption

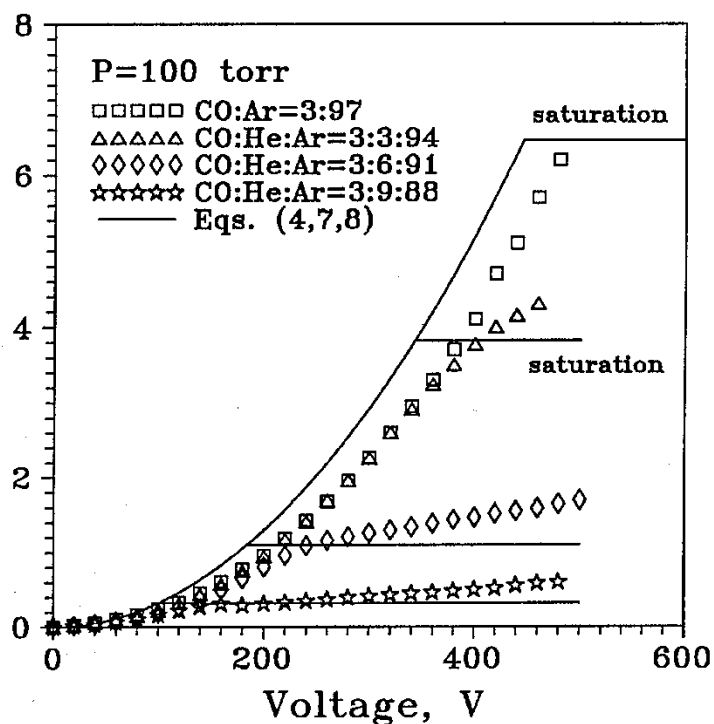


**Electron density:**

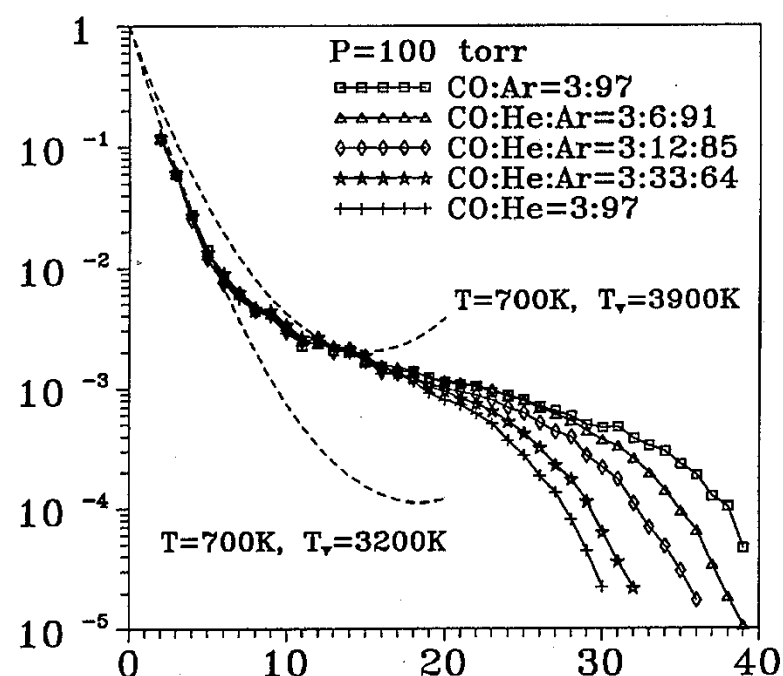
$$n_e \cong 10^{11} \text{ cm}^{-3}$$



Current,  $\mu\text{A}$



Current-voltage characteristics of the DC Thomson discharge at different helium partial pressures



Vibrational distribution functions of carbon monoxide at different helium partial pressures

# Optically Pumped Plasmas: Kinetics Studies

## Coupling of Vibrational Populations with Free Electrons

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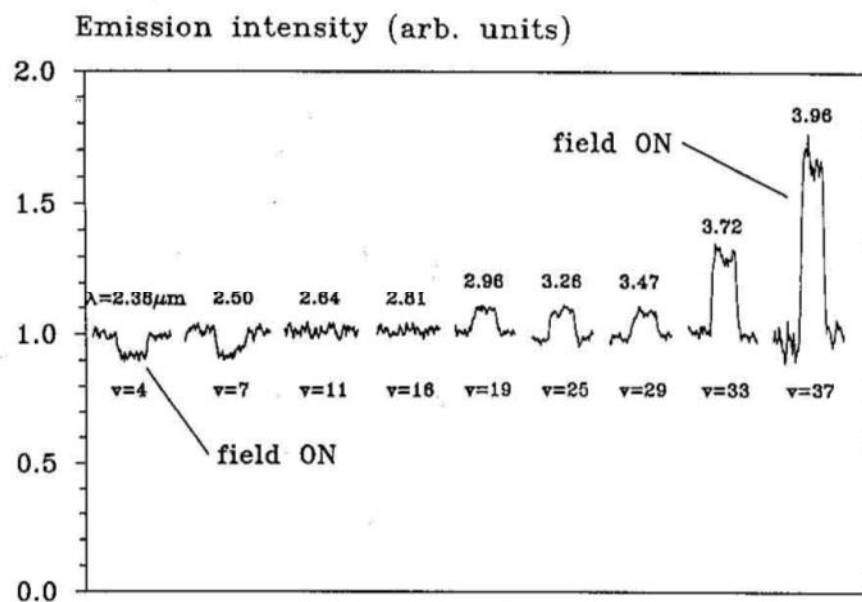


Fig. 11. Electric field influence on different CO vibrational level populations.

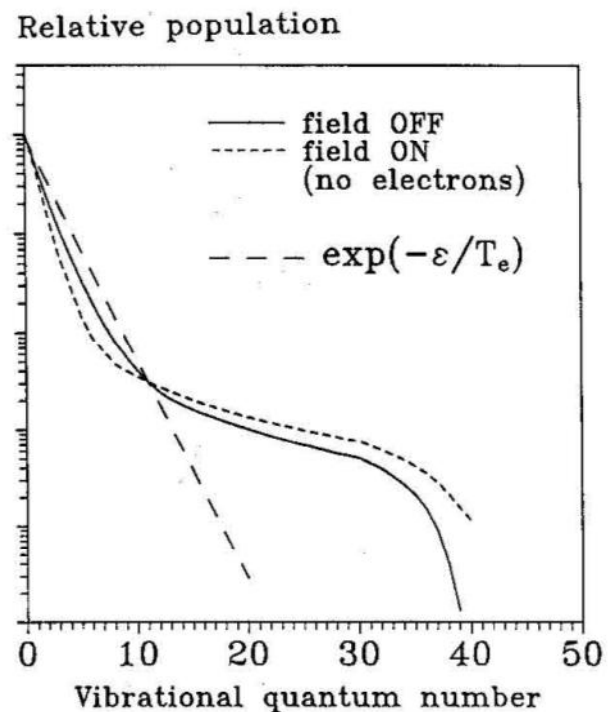


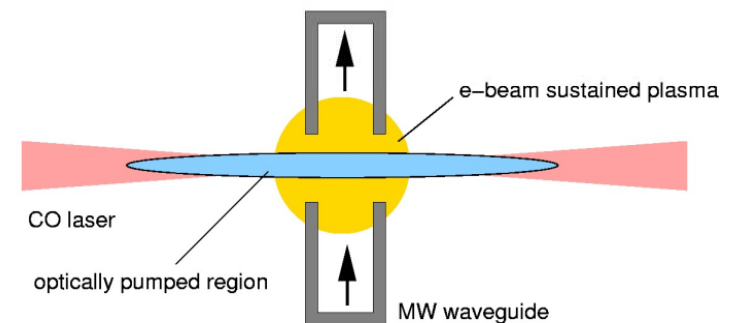
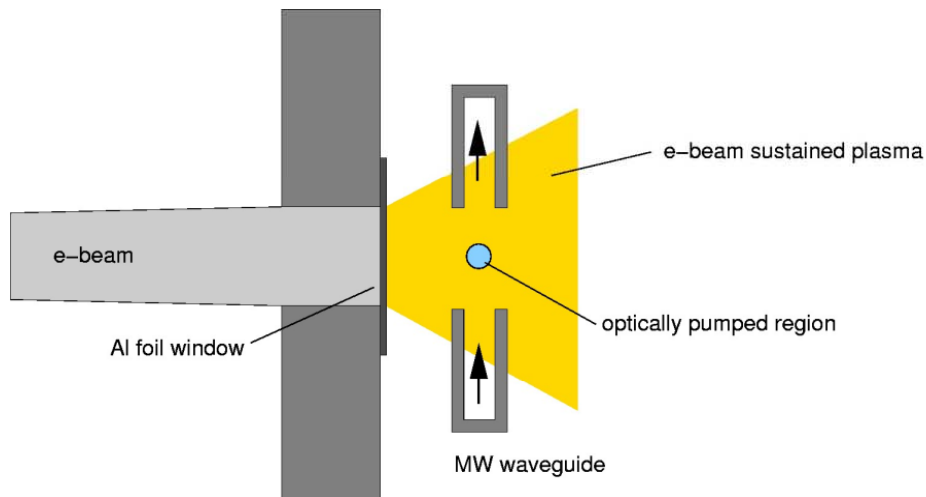
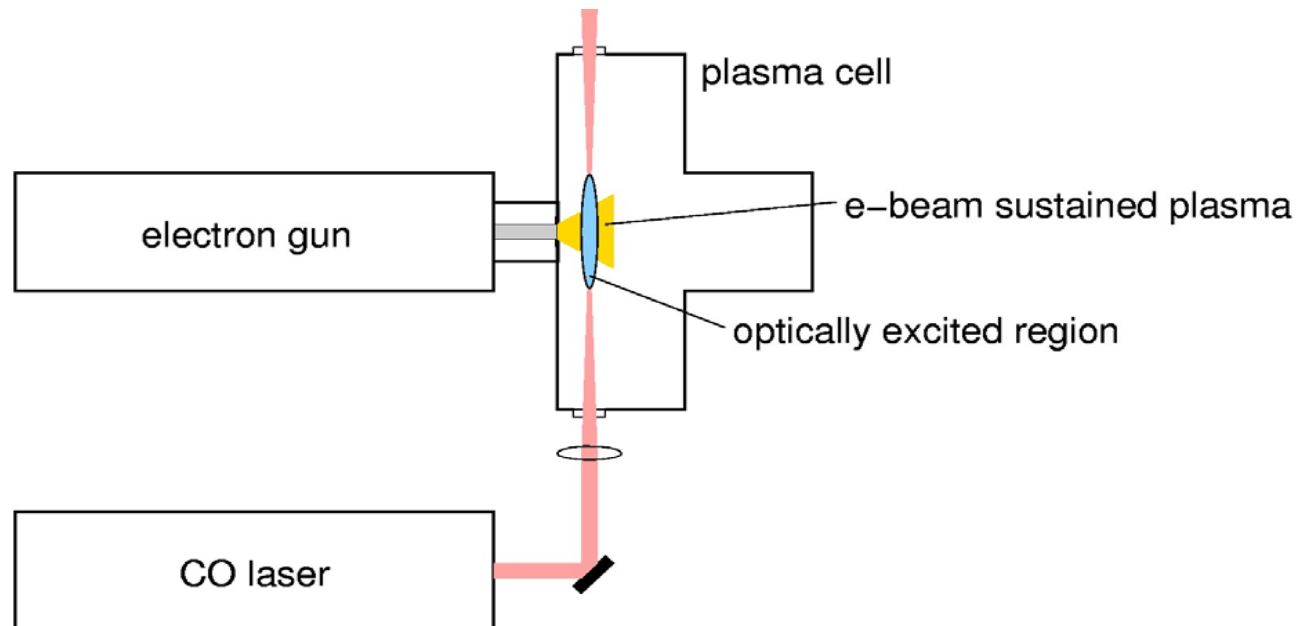
Fig. 12. Qualitative demonstration of the  $V \rightarrow e \rightarrow V - \Delta V$  effect (strongly exaggerated).  $\epsilon$ : electron energy;  $T_e$ : electron temperature.

# Optically Pumped Plasmas: Kinetics Studies

## Measurements of Electron Density Decay (by microwave attn) for E-beam Created Plasma – Experimental Schematic

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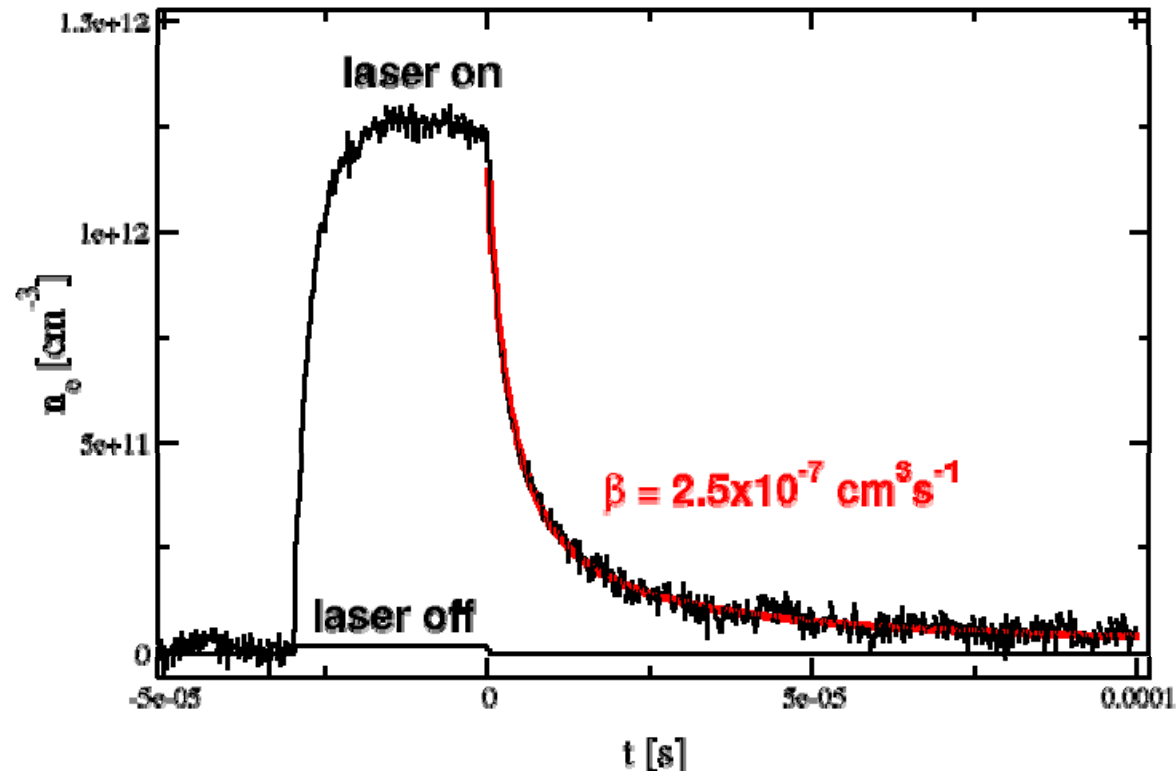


## Optically Pumped Plasmas: Kinetics Studies

### Measurements of Electron Density Decay (by microwave attn) for E-beam Created Plasma – Inhibition of Electron Loss Processes in Cold Air

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Plasma Power Consumption\_at  $10^{13}$  Electrons/ $\text{cm}^3$  in 1 Atm, 560 K Air:

#### Electron-Ion Recombination

$$\beta = 2.5 \times 10^{-7} \text{ cm}^3 \text{ sec}^{-1}$$

Nonequilibrium: 56 W  $\text{cm}^{-3}$

Equilibrium: 450 W  $\text{cm}^{-3}$

#### Electron Attachment to $\text{O}_2$

$$k_{\text{eff}} = 2.3 \times 10^{-33} \text{ cm}^6 \text{ sec}^{-1}$$

Nonequilibrium: 1.2 W  $\text{cm}^{-3}$

Equilibrium: 1.4 kW  $\text{cm}^{-3}$



# Optically Pumped Plasmas: Kinetics Studies

## Inhibition of Electron Loss Processes in Cold Air

### Influence of Temperature on O<sub>2</sub> Attachment and Detachment Rates

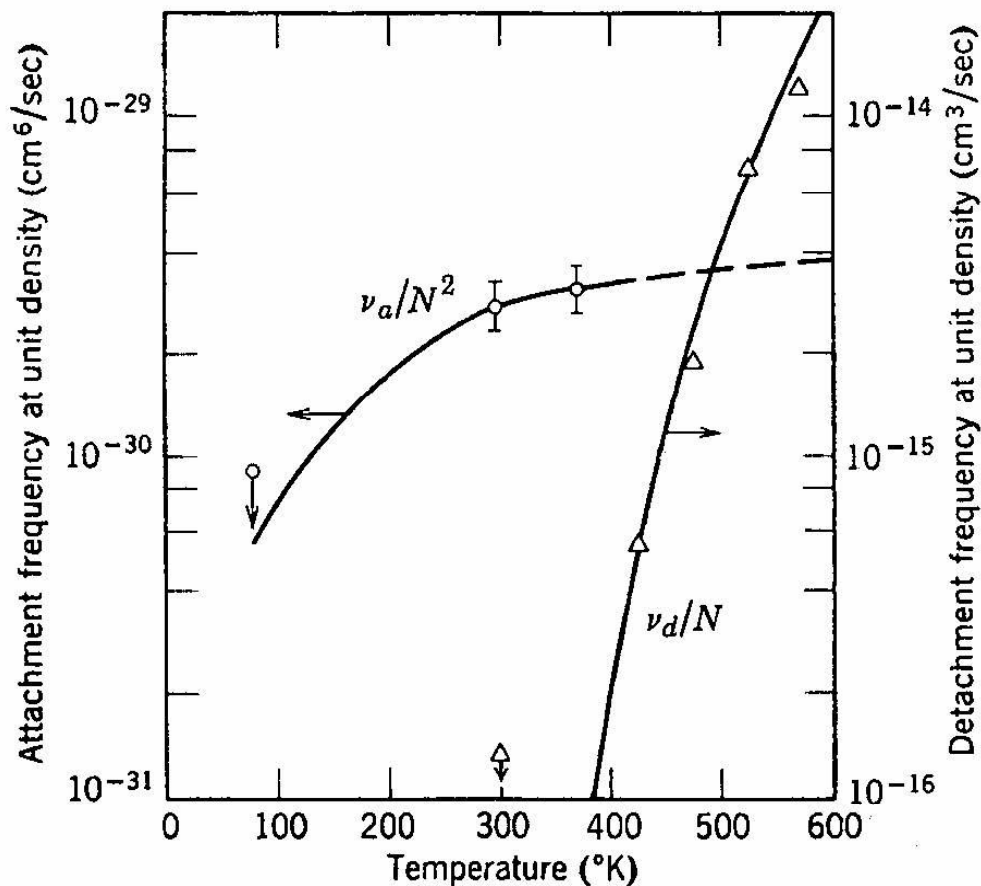
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**Attachment is weakly dependent upon heavy species temperature  
whereas detachment is highly dependent**



**O<sub>2</sub> Electron Affinity ~0.43 eV  
(~Two Vibrational Quanta)**

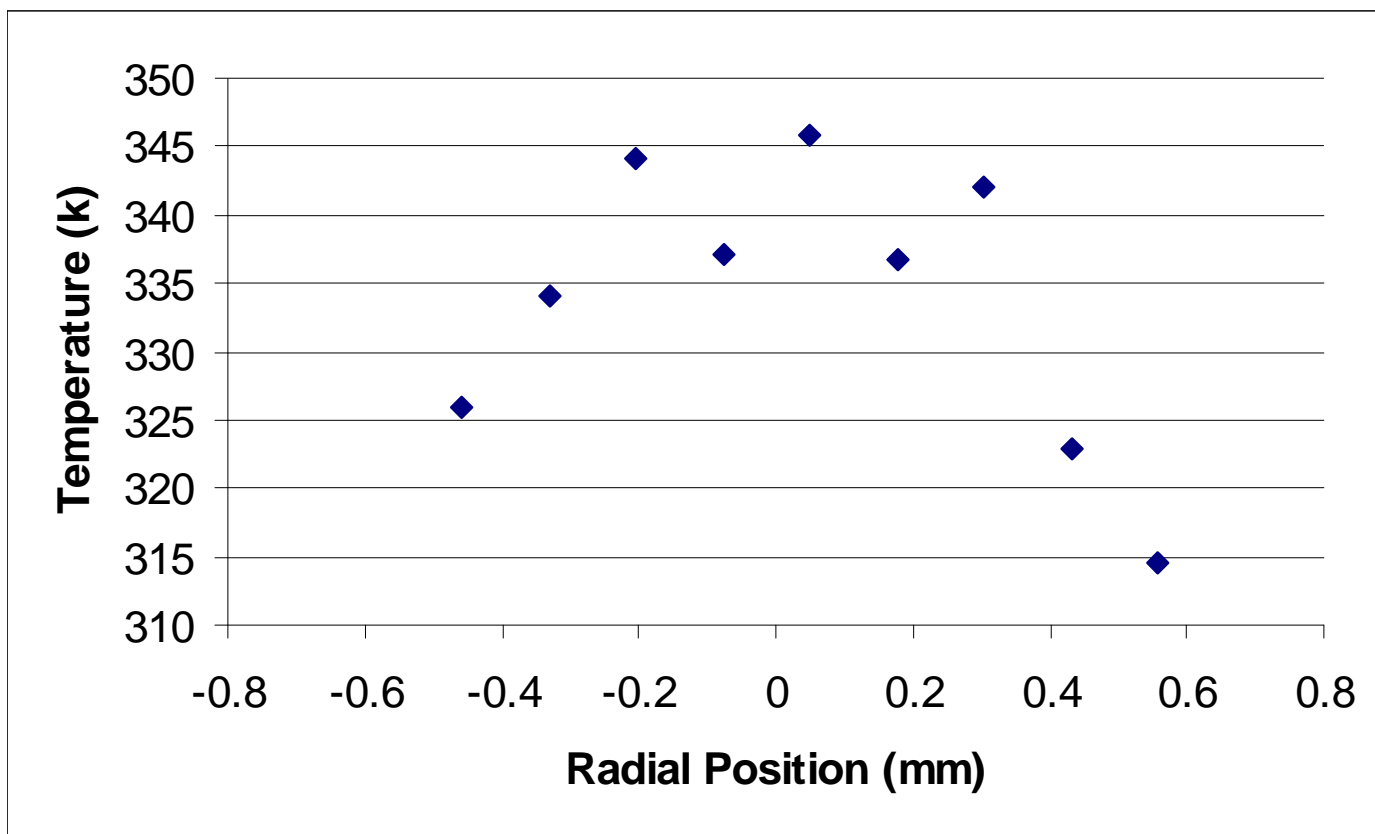


**Optically Pumped Plasmas: Kinetics Studies**  
**Inhibition of Electron Loss Processes in Cold Air**  
**Radially-Resolved Filtered Rotational Raman Temperature Measurement**

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**200/18/35 N<sub>2</sub>/CO/O<sub>2</sub> – Optically Pumped**  
**(Diameter of Vibrationally Excited Region ~ 1 mm)**



## Gas Dynamic Flows: Vibrational mode nonequilibrium in a supersonic nozzle expansion

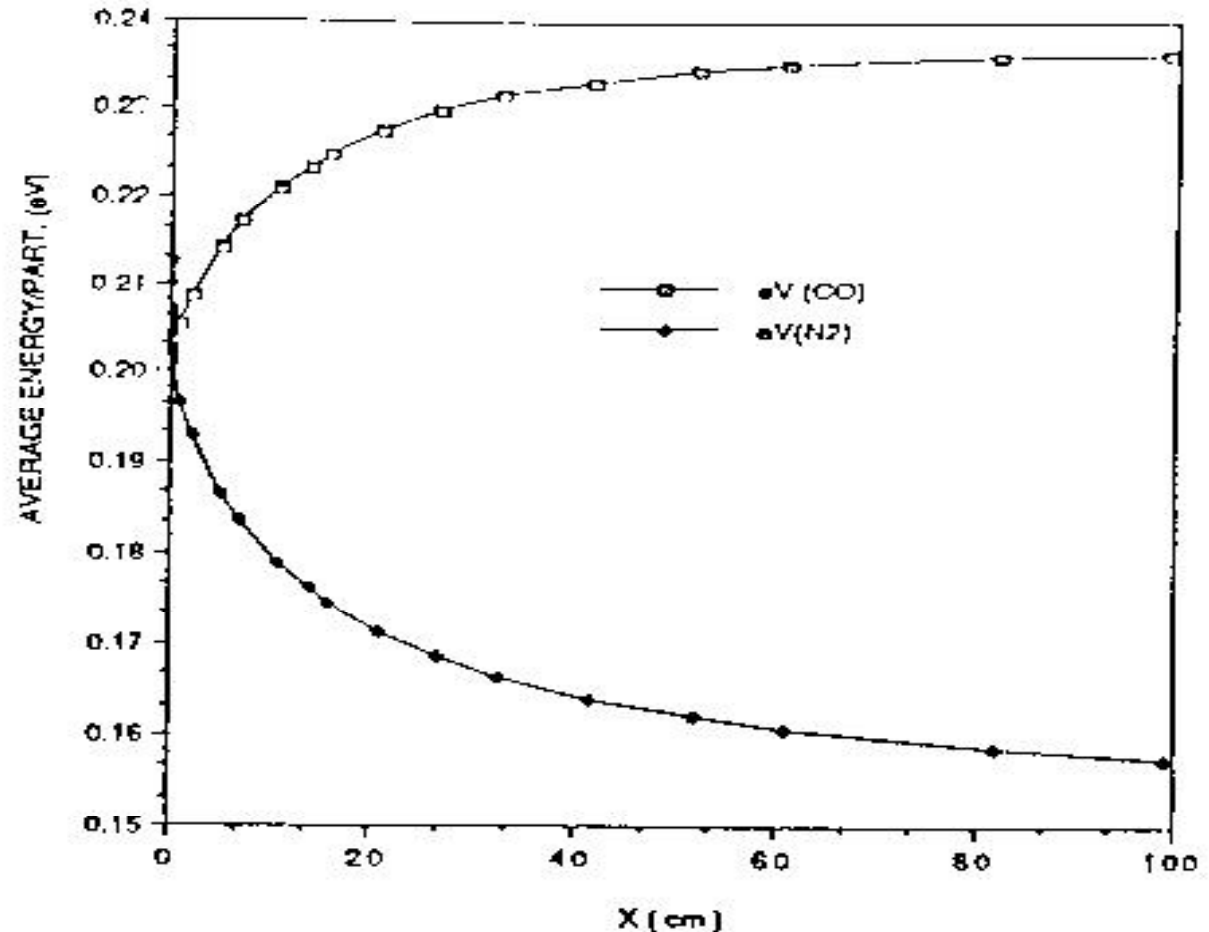
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Average energy per  
molecule in electron  
Volts.

(0.1 eV = 1,161 °K)

Expansion of a gas  
mixture of 20% CO,  
20% N<sub>2</sub>, 60% Ar,  
Stagnation Pressure =  
100 atm.  
Stagnation Temperature =  
2,000 °K.  
15 ° Half Angle Nozzle,  
Expands to M = 10 at  
100 cm downstream of throat



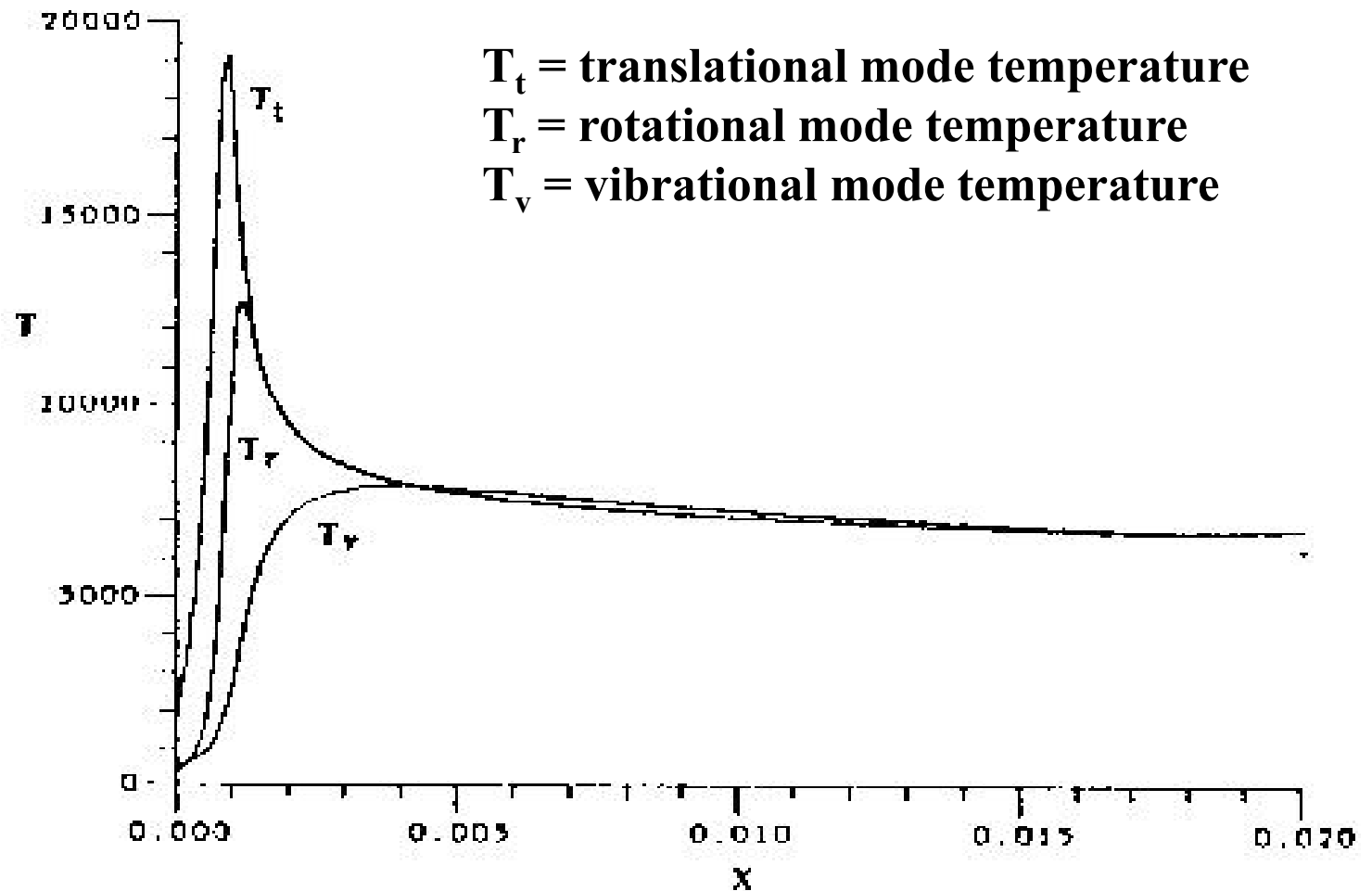
Distance from nozzle throat, in centimeters

# Gas Dynamic Flows: Thermal mode nonequilibrium behind a hypersonic shock wave

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Mode  
Temperature  
Degrees K

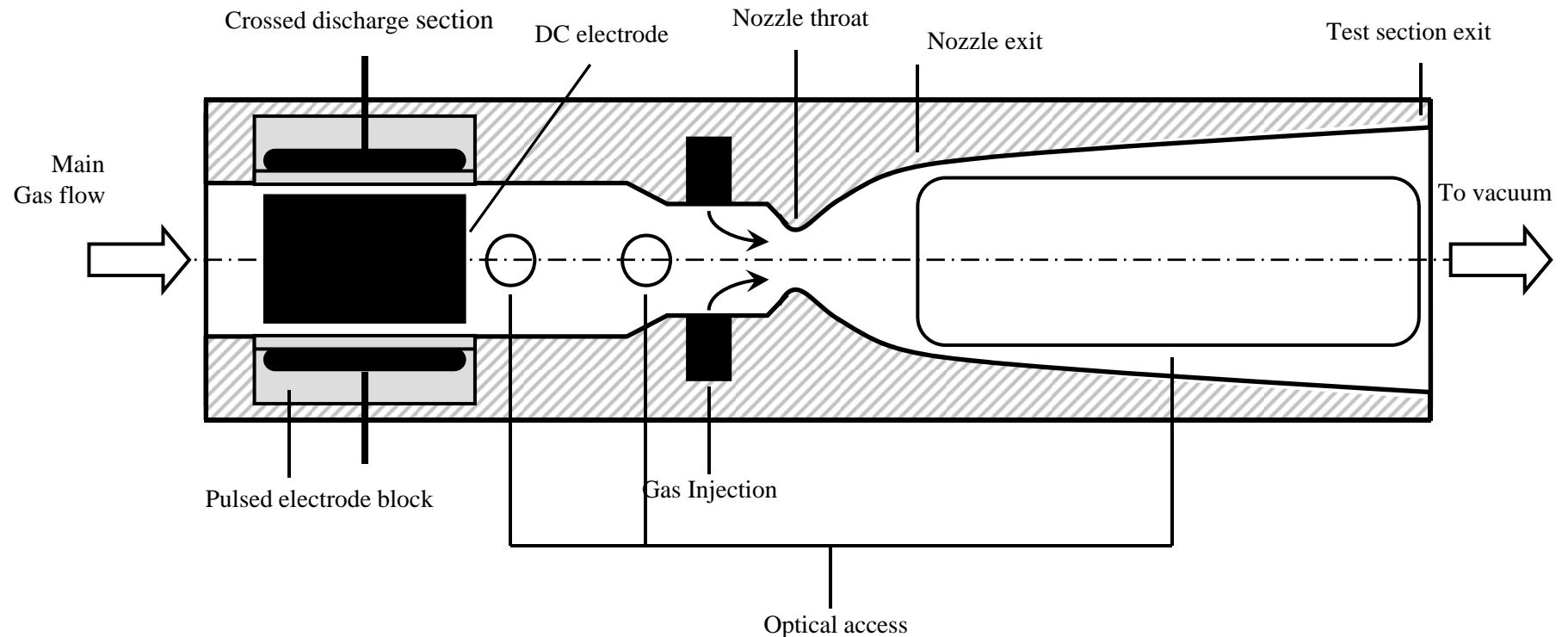


Distance behind shock front, in meters

# Plasma Wind Tunnels: A supersonic tunnel with energy loading of selected internal states in the plenum

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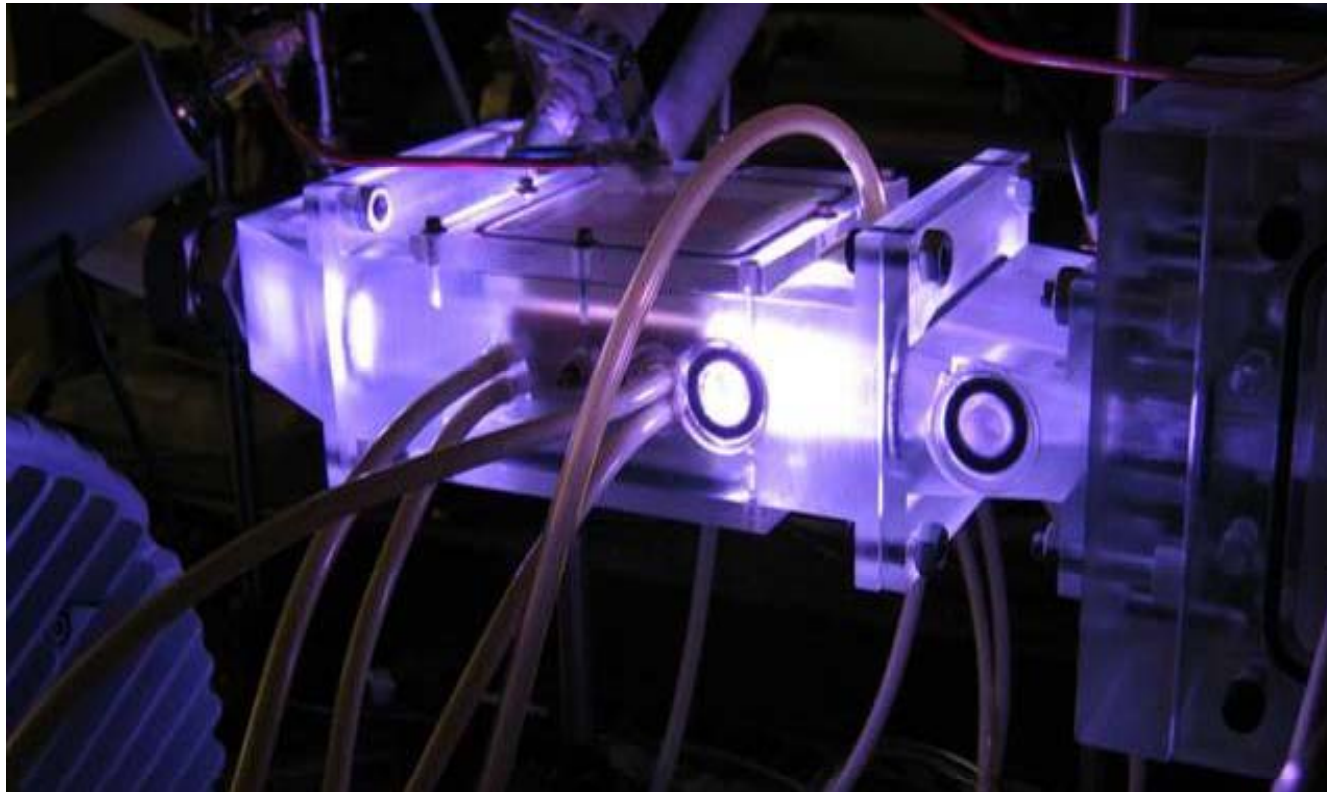


**Schematic of the wind tunnel  
discharge section / nozzle / test section assembly**

## **Plasma Wind Tunnels: A supersonic tunnel with energy loading of selected internal states – photo of plenum in operation**

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**Photograph of a discharge in the nozzle plenum. Dimensions 4x4x10 cm.**

**A uniform, cold gas fills the plenum;**

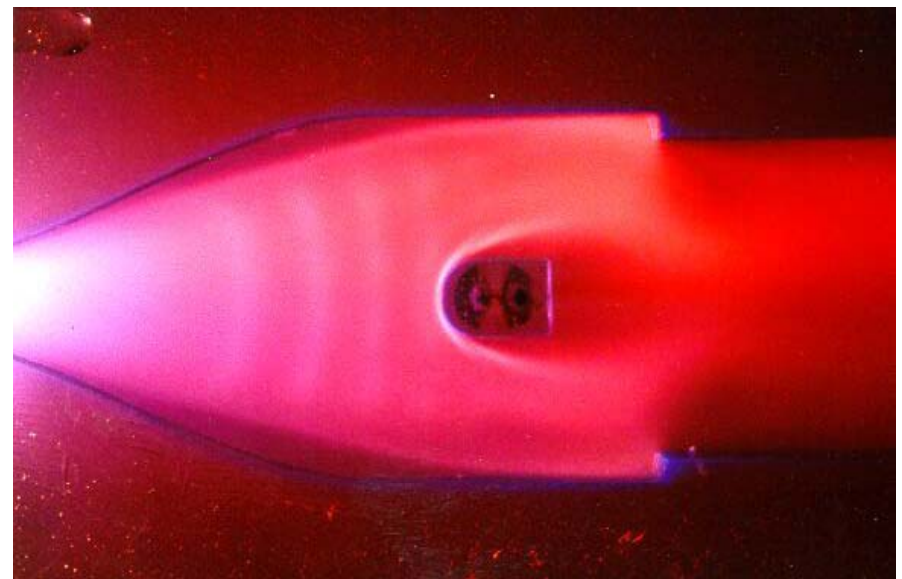
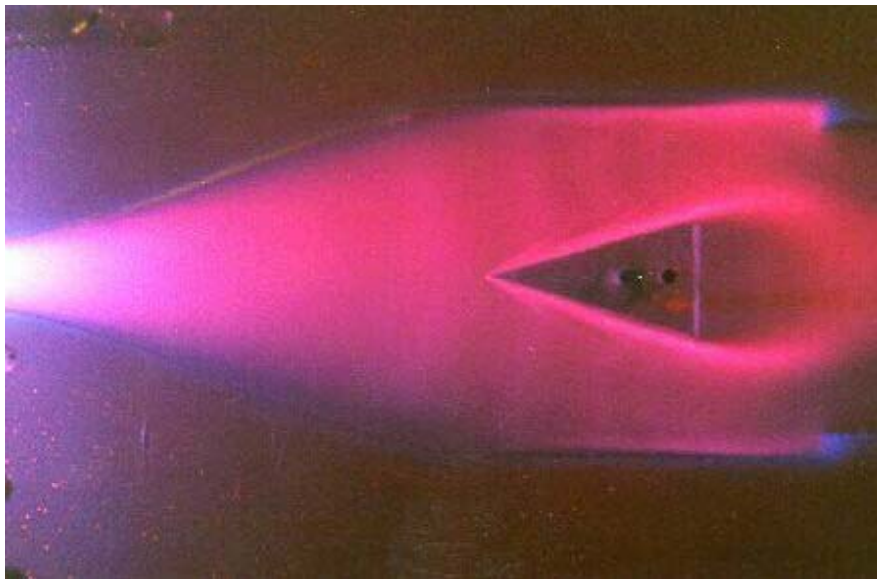
**O<sub>2</sub>/He mixture shown, similar uniformity for air; P<sub>0</sub> to 1 atm.**



**Plasma Wind Tunnels: A supersonic tunnel with energy loading  
of selected internal states – objects in a 2-D,  $M = 3$  test section**

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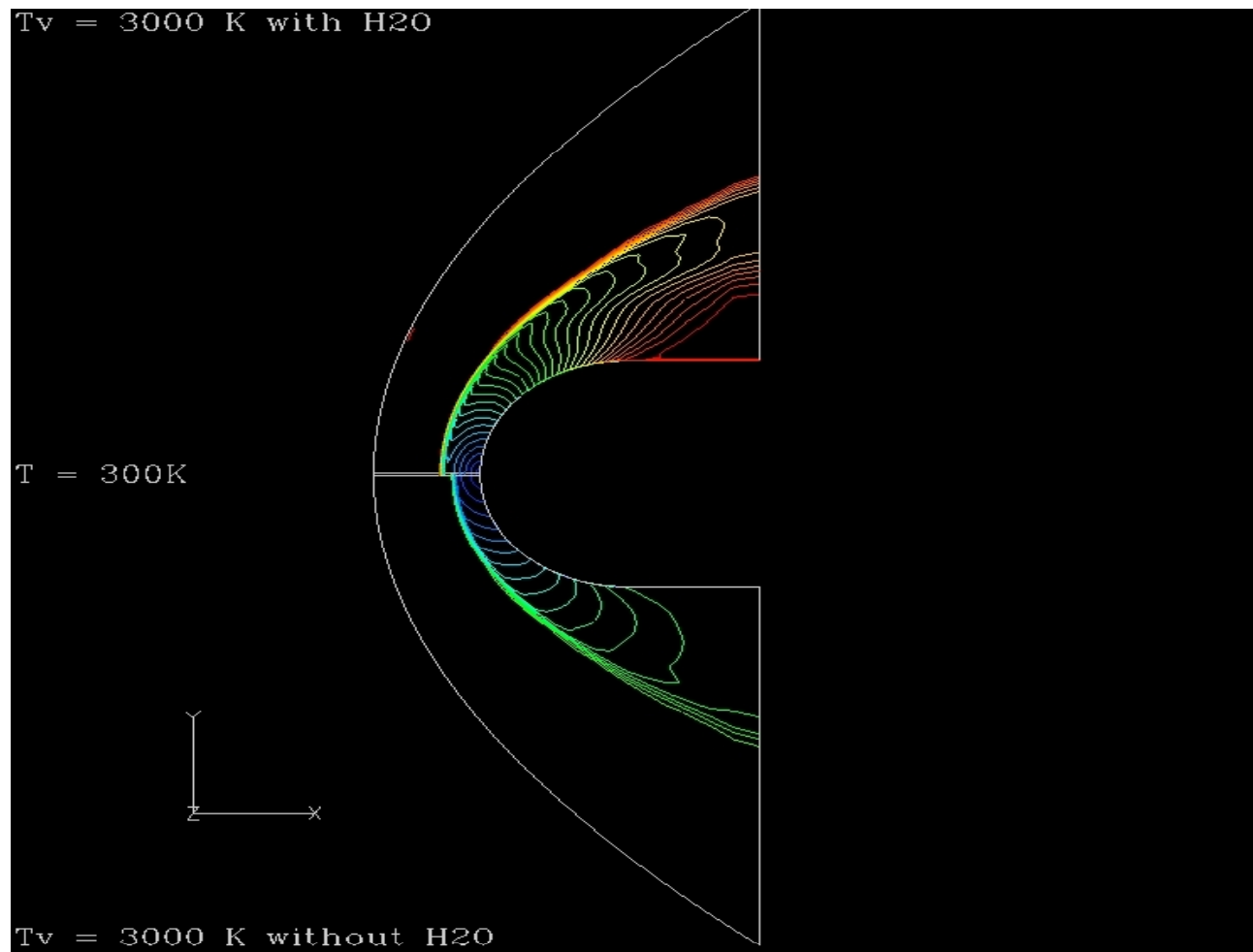
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**Plasma Wind Tunnels: Calculated  $M=4$  Flow over a 0.5 cm Dia semicylinder. Vibrationally Excited  $N_2$  Expanding from 1 atm.**

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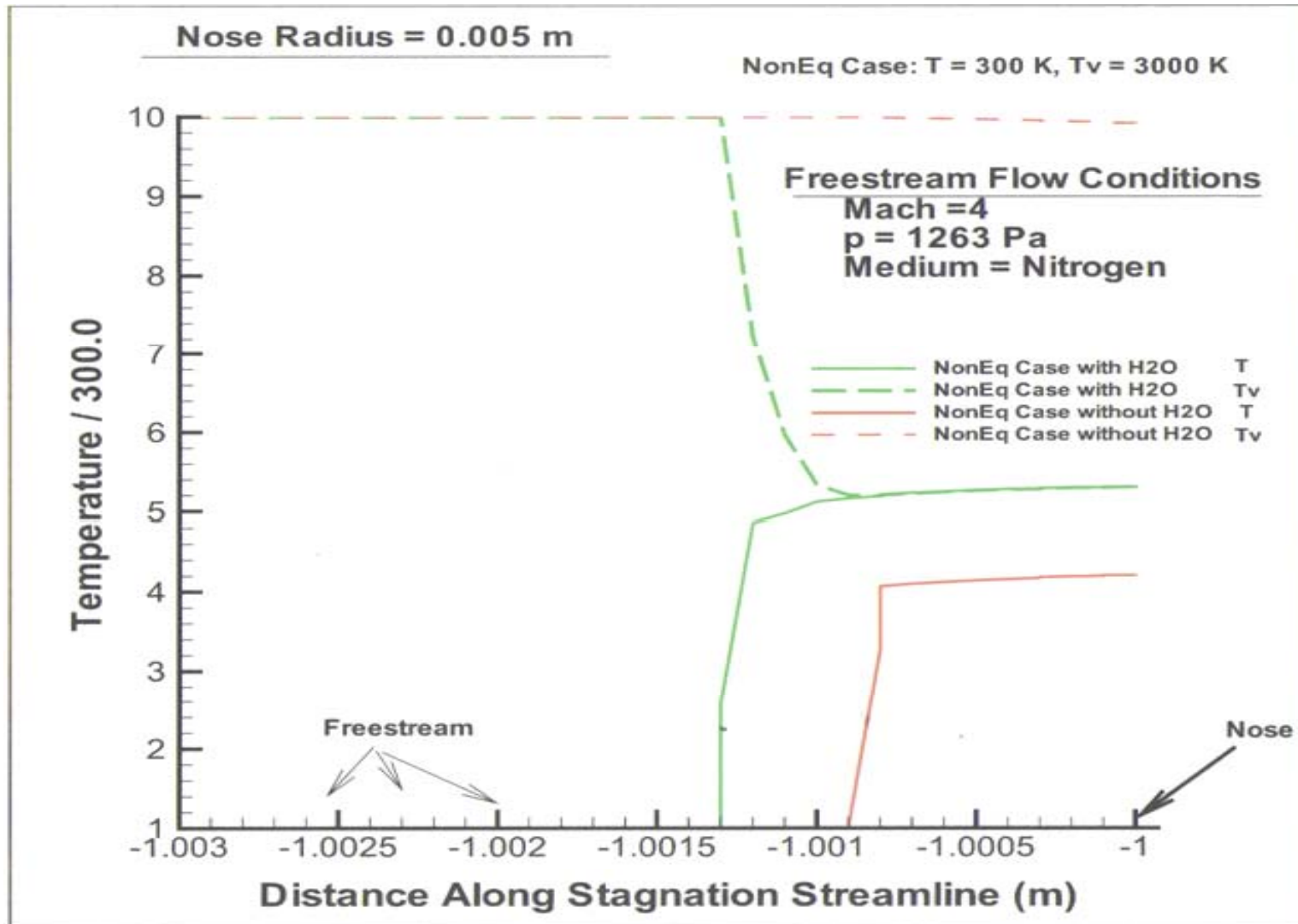
From E. Josuyla,  
AFRL

# Plasma Wind Tunnels: Calculated M =4 Flow over a 0.5 cm Dia semicylinder. Vibrationally Excited N<sub>2</sub> Expanding from 1 atm.

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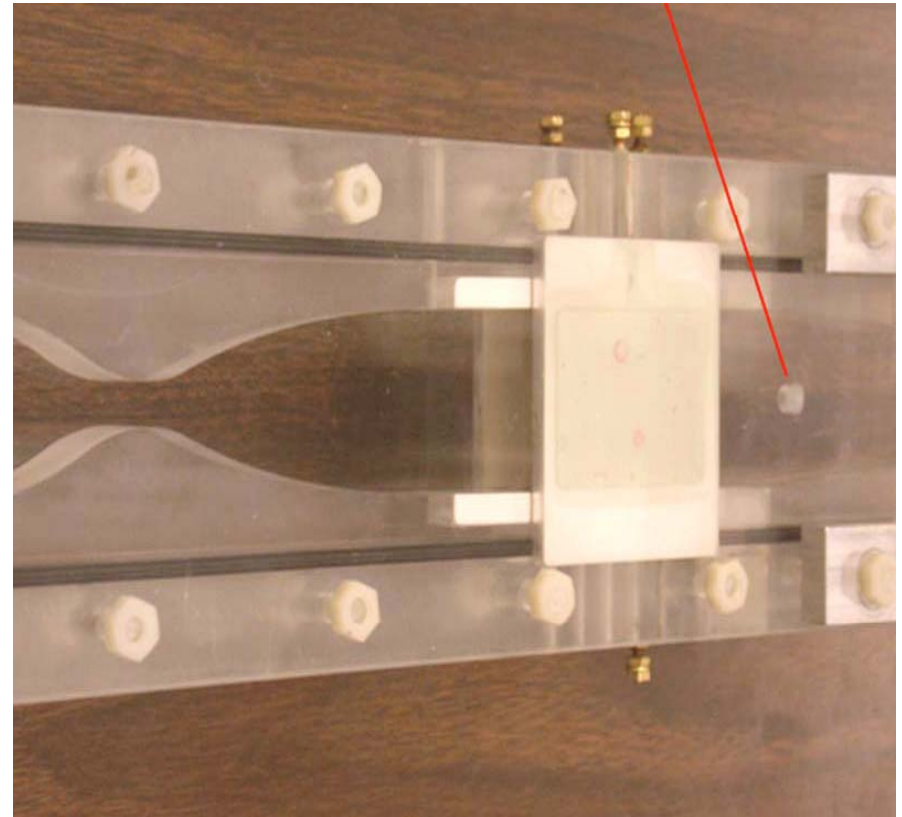
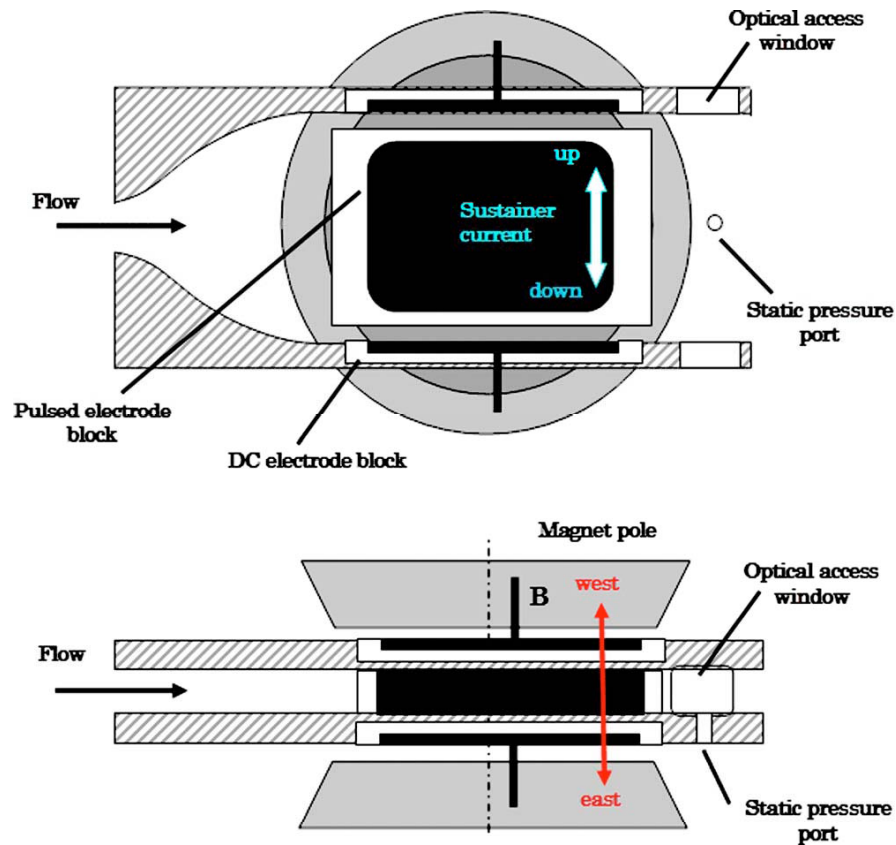
From E. Josuyl, AFRL



# Plasma Wind Tunnels: An $M = 4$ MHD Tunnel, with transverse pre-ionized discharge and a 2 Tesla Field in test section

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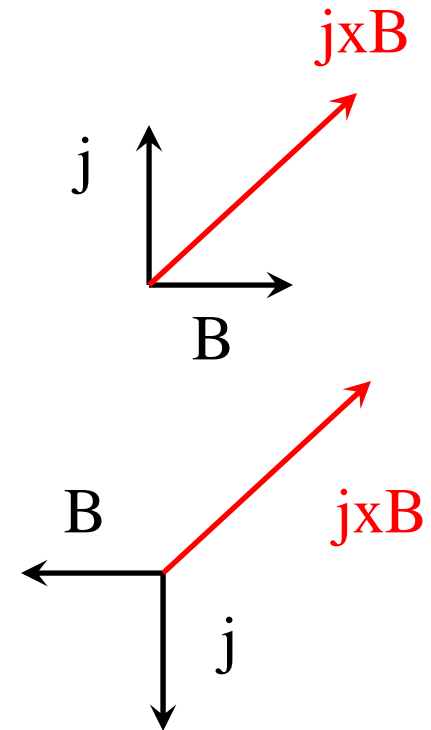
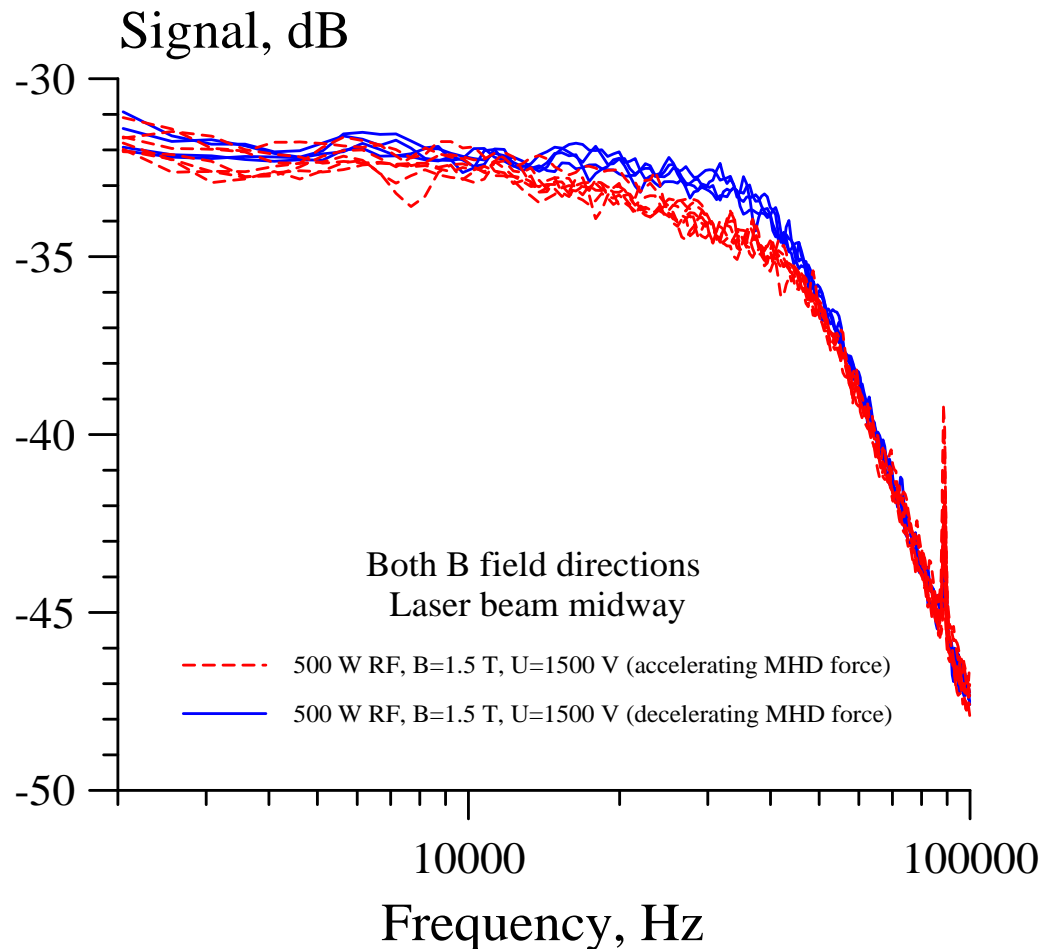
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# Plasma Wind Tunnels: An $M = 4$ MHD Tunnel. Lorentz force effect on turbulent boundary layer density fluctuation spectra

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**Nitrogen,  $M=3$ ,  $P_0=1/3$  atm**

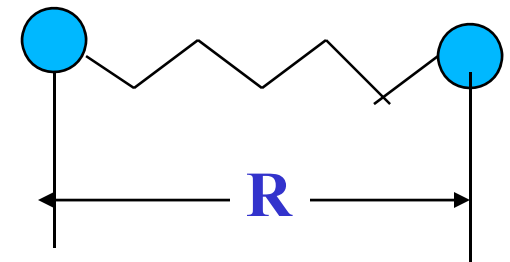
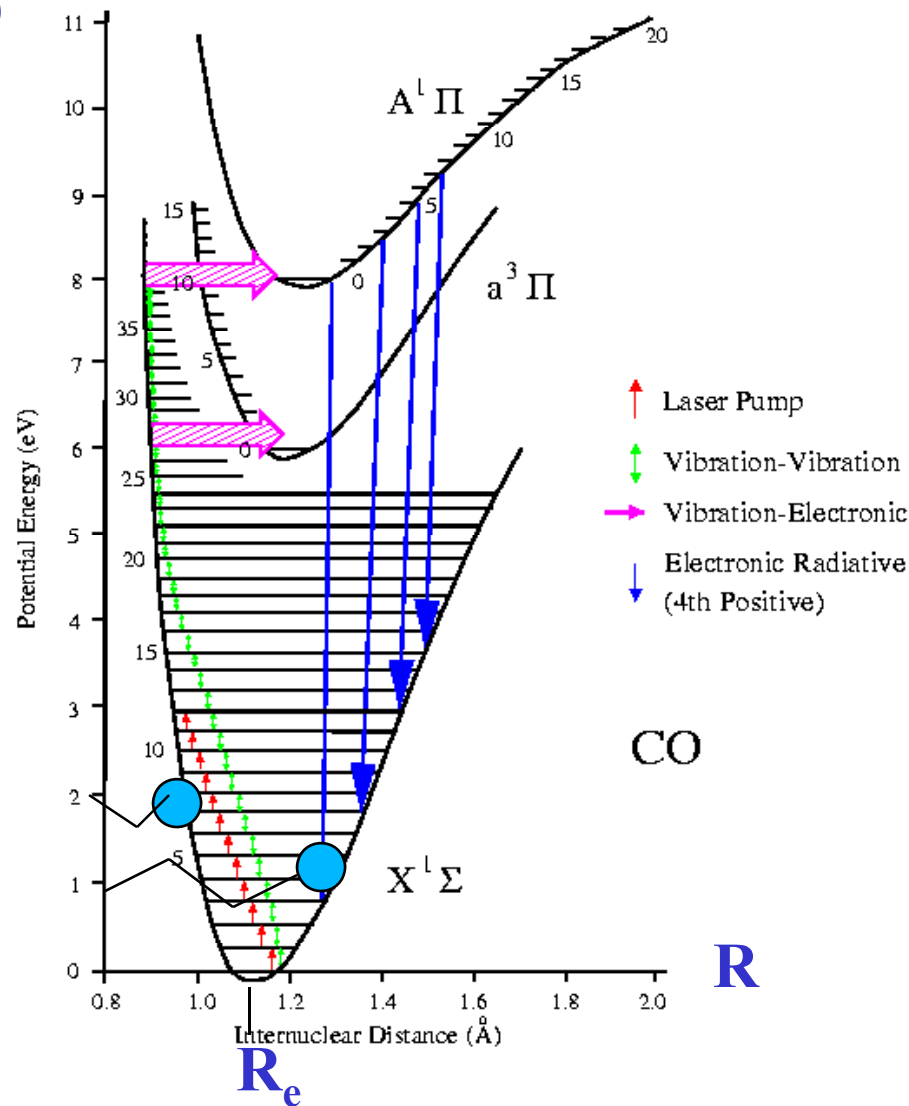
**Both accelerating and decelerating Lorentz force are created for two possible combinations of  $B$  and  $E$  fields, as shown**

# CO potential

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$V(R)$



$R$  is the separation of the C and O nuclei.  
 $R_e$  is the equilibrium (nonvibrating) separation.

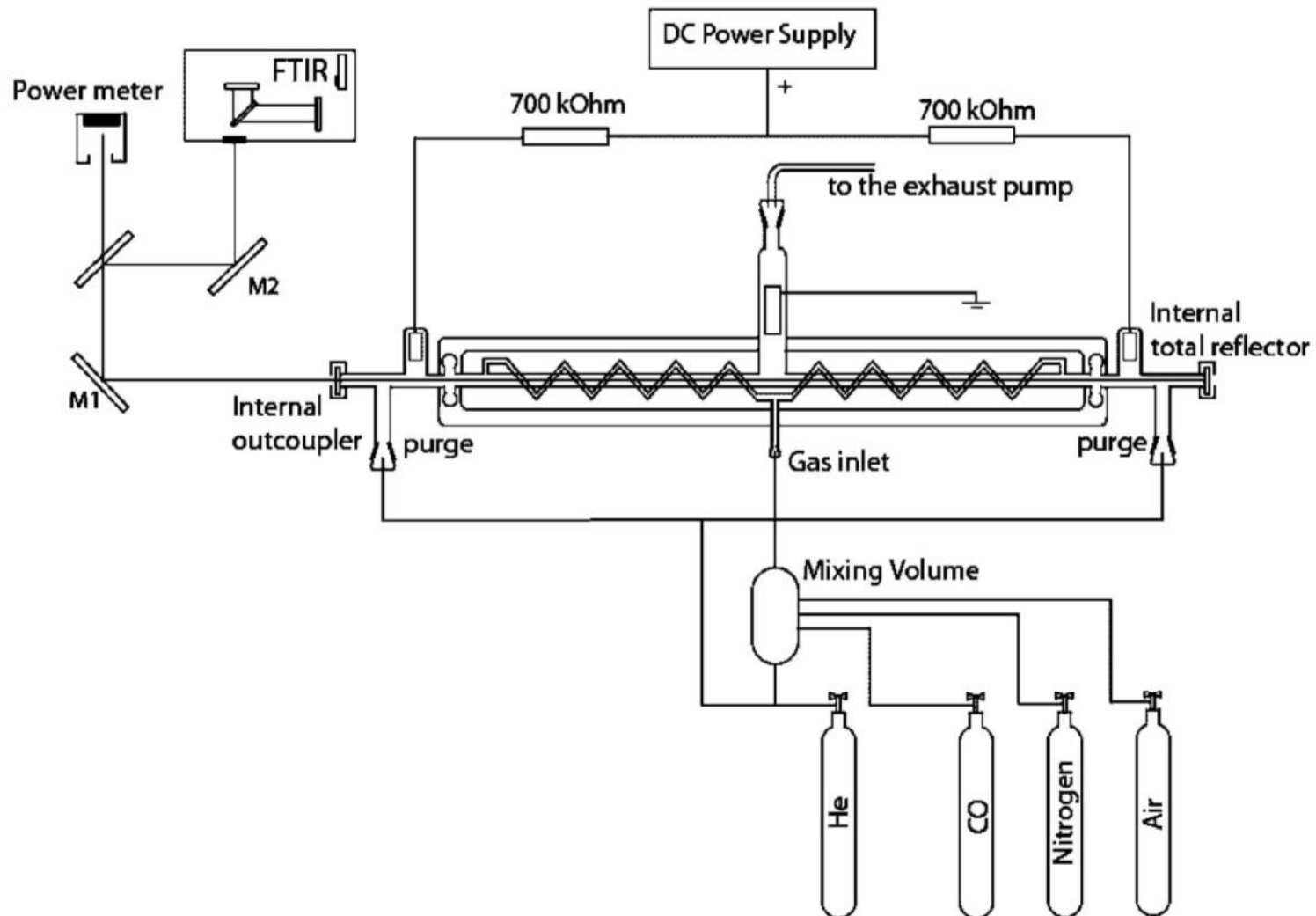
$V(R) = 1/2 K(R - R_e)^2$  is the potential energy stored in the oscillator



# Gas Lasers: Carbon Monoxide Electric Discharge Laser - Schematic

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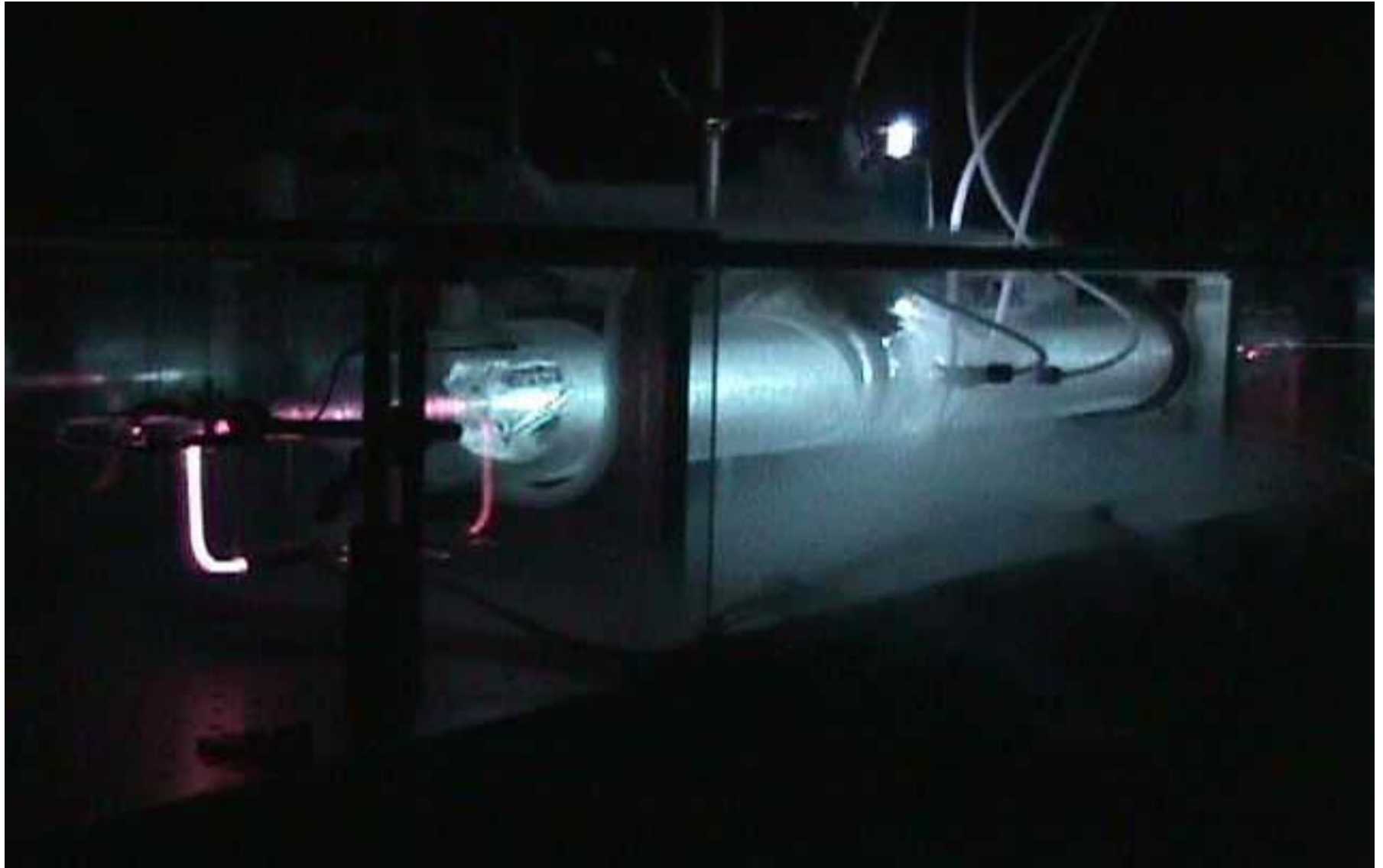
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## **Gas Lasers: Carbon Monoxide Electric Discharge Laser – Photo during operation**

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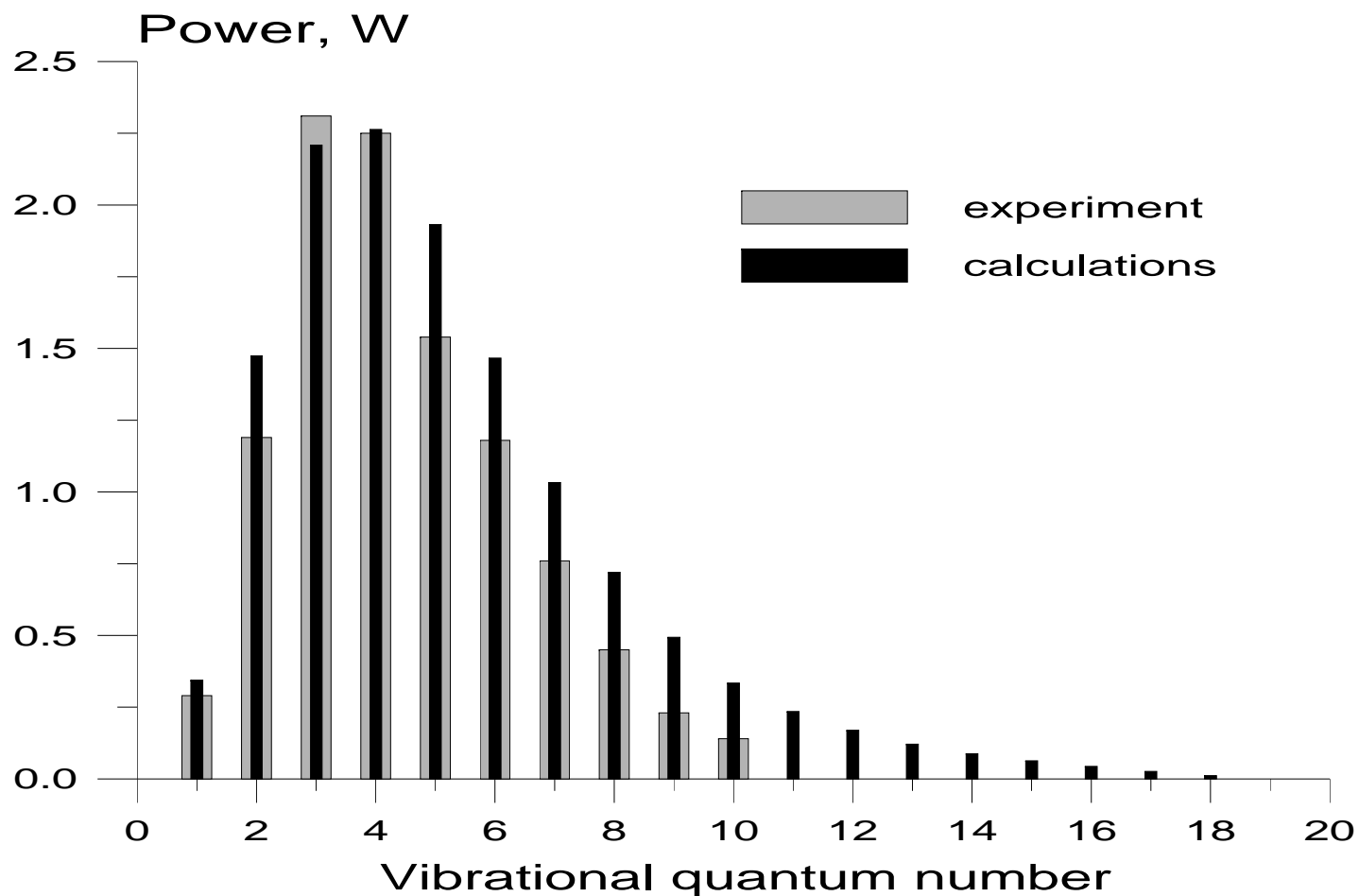
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# Gas Lasers: Carbon Monoxide Electric Discharge Laser – Kinetic Modeling of CO Fundamental Band Laser, Comparison with OSU Laser Performance

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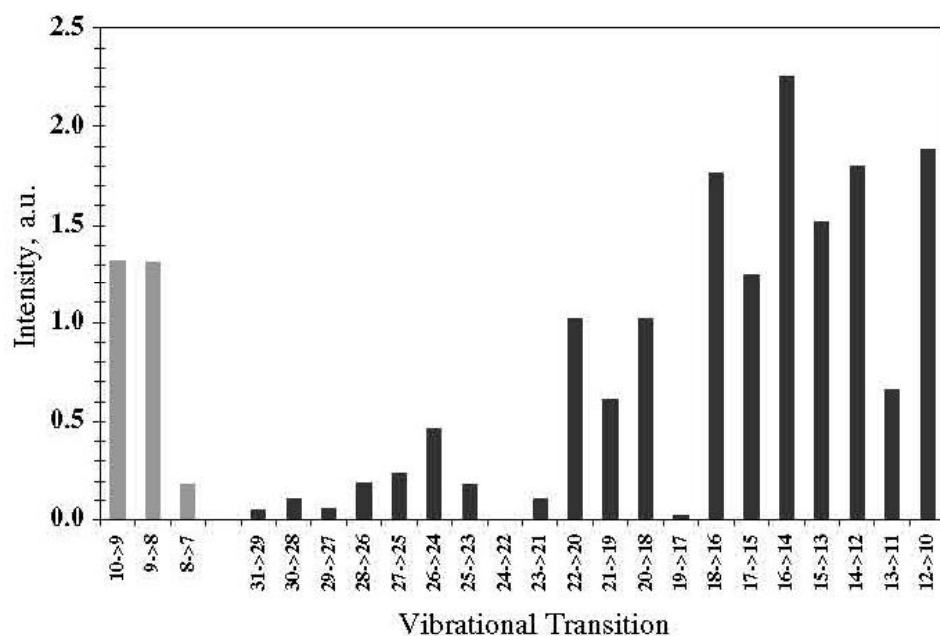
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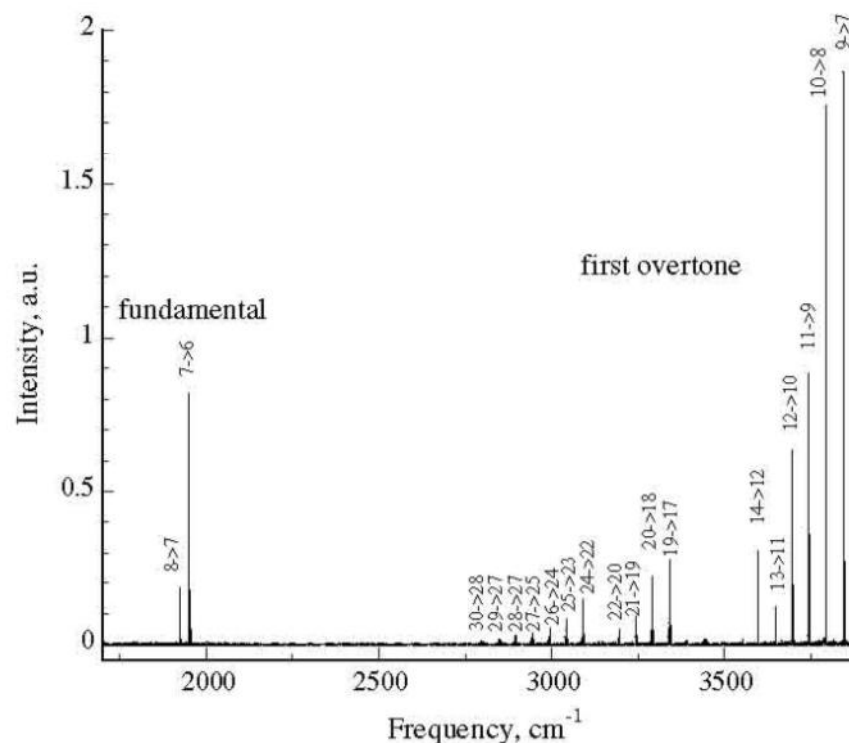
# Gas Lasers: Carbon Monoxide Electric Discharge Laser – Kinetic Modeling of CO Overtone Band Laser, Comparison with OSU Laser Performance

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**Theoretically Predicted Output Line Intensities**

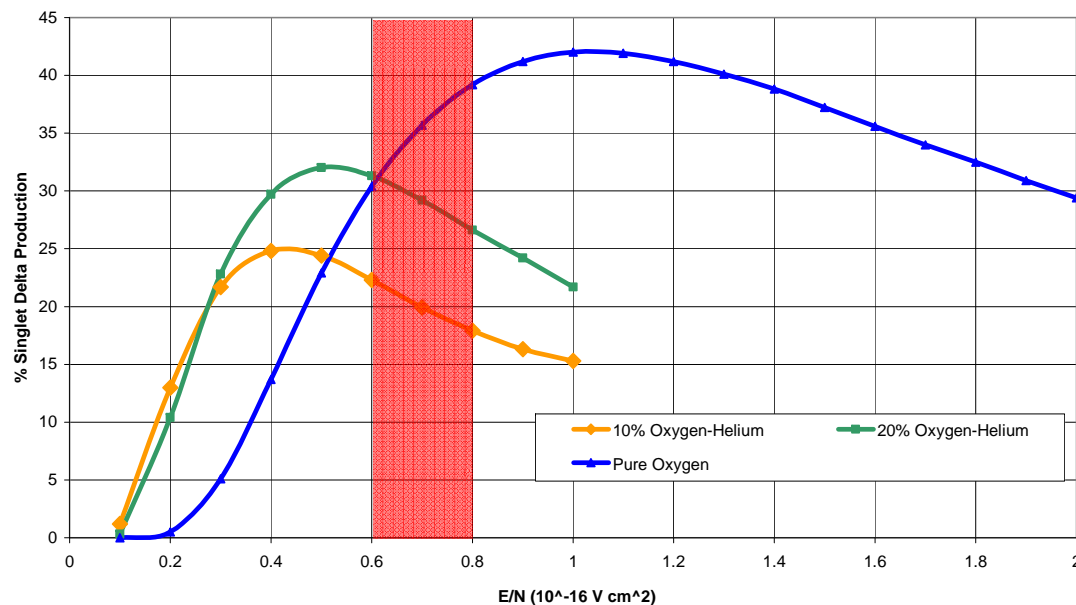


**Experimentally Measured  
Output Line Intensities**

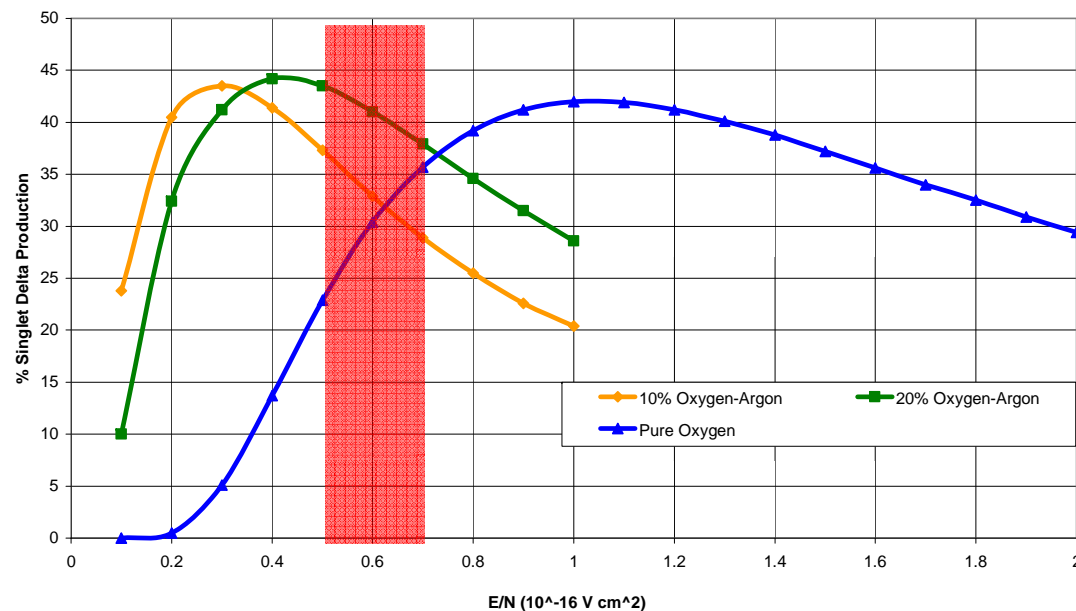
# Gas Lasers: An Electric Discharge-Excited Oxygen-Iodine Laser (DOIL) – Boltzmann equation solver results: electron energy balance

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**O<sub>2</sub>-He**



**O<sub>2</sub>-Ar**

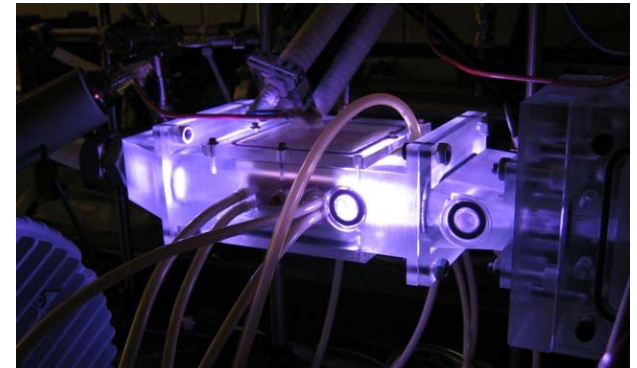
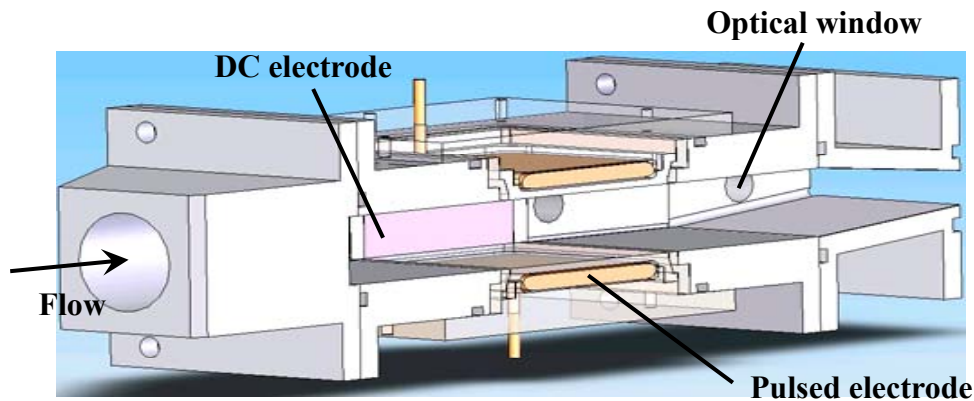
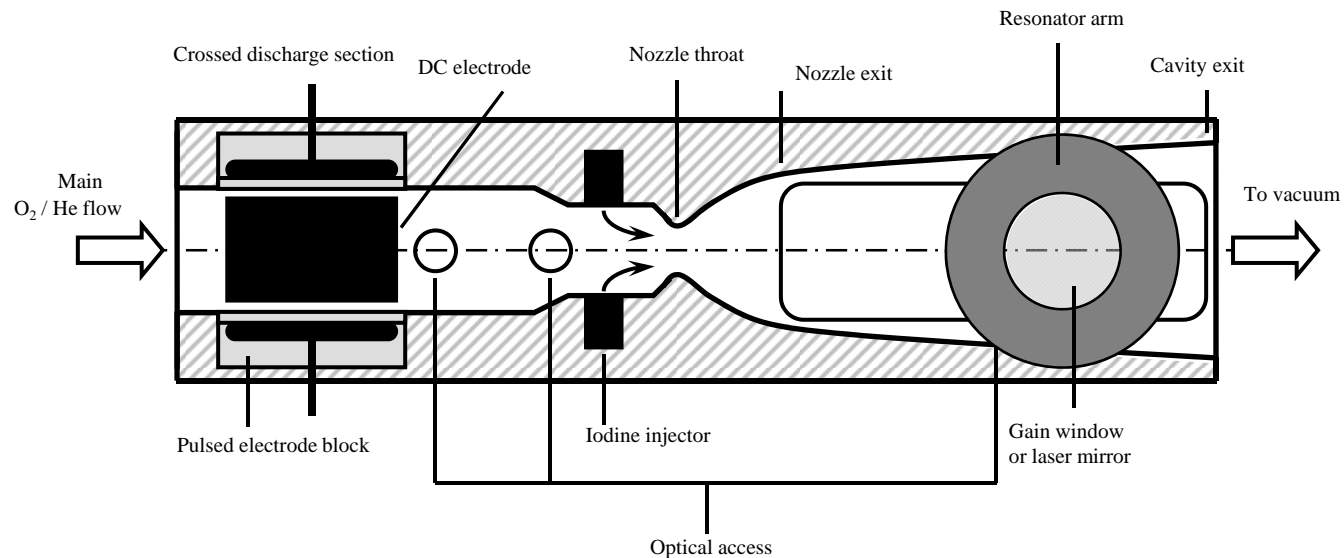
**Highest estimated  
O<sub>2</sub>(<sup>1</sup>Δ) yield achieved  
so far ~10 %**

## Gas Lasers: An Electric Discharge-Excited Oxygen-Iodine Laser (DOIL) Schematic of Electric Discharge SDO Generator

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**Advantages of transverse vs. axial discharge:** Large volume, stable at high pressures / powers, rapid convective cooling vs. slow wall cooling. Used in most high power electrically excited lasers.



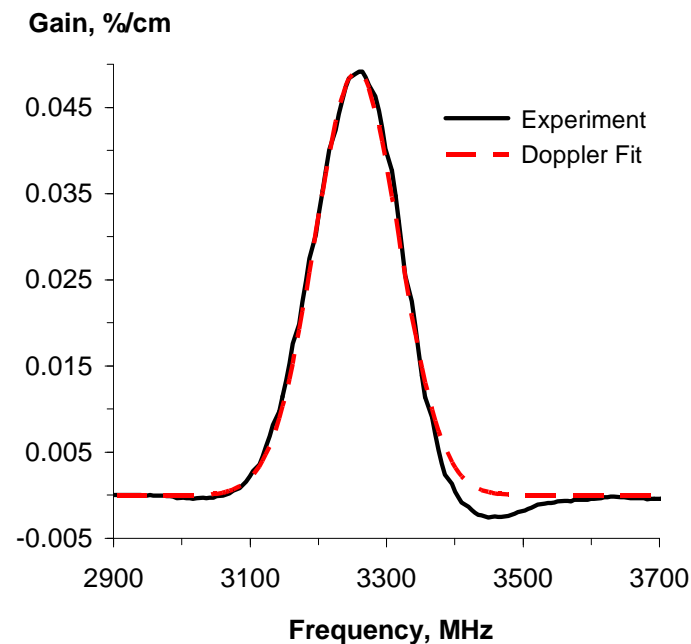
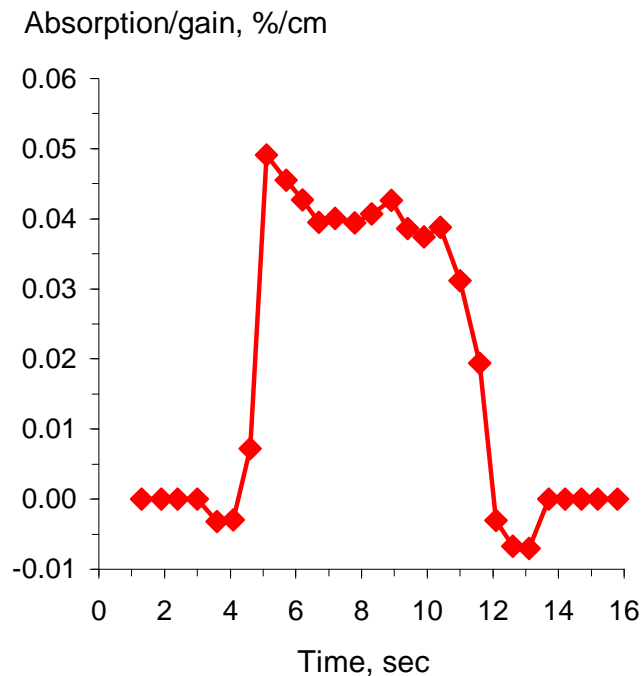
Discharge volume 50 cm<sup>3</sup>, pressures up to 460 torr, flow rate up to 0.1 mole/sec (O<sub>2</sub>), 1 mole/sec (He)

# Gas Lasers: An Electric Discharge-Excited Oxygen-Iodine Laser (DOIL)

Gain at Optimized Conditions: 0.12 %/cm at T=100 K (Currently!)

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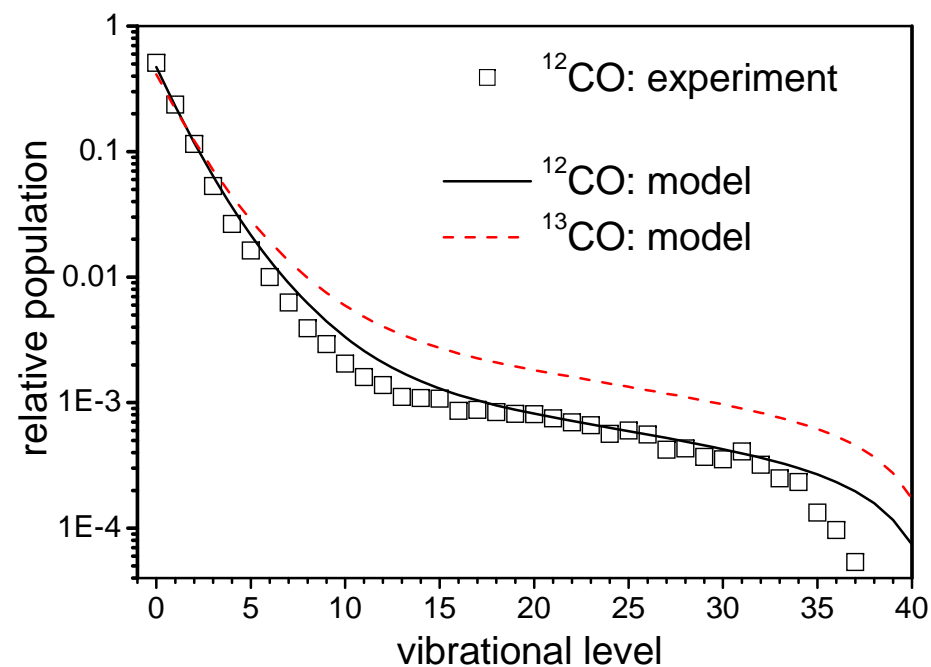
**Adding NO: major discharge stabilization factor**

**Discharge power increased from 1.9 kW to 2.4 kW**

**$P_0=107$  torr, 15% O<sub>2</sub> - He flow, NO 0.2 mmole/sec (550 ppm),  $\nu=34$  kHz,  $U_{PS}=3.1$  kV,  $I=1.54$  A, discharge power 2.4 kW, I<sub>2</sub> 70  $\mu$ mole/sec (190 ppm).**

**Gain may be limited by I<sub>2</sub> flow rate**





Vibrationally excited CO prepared in cell reacts according to:

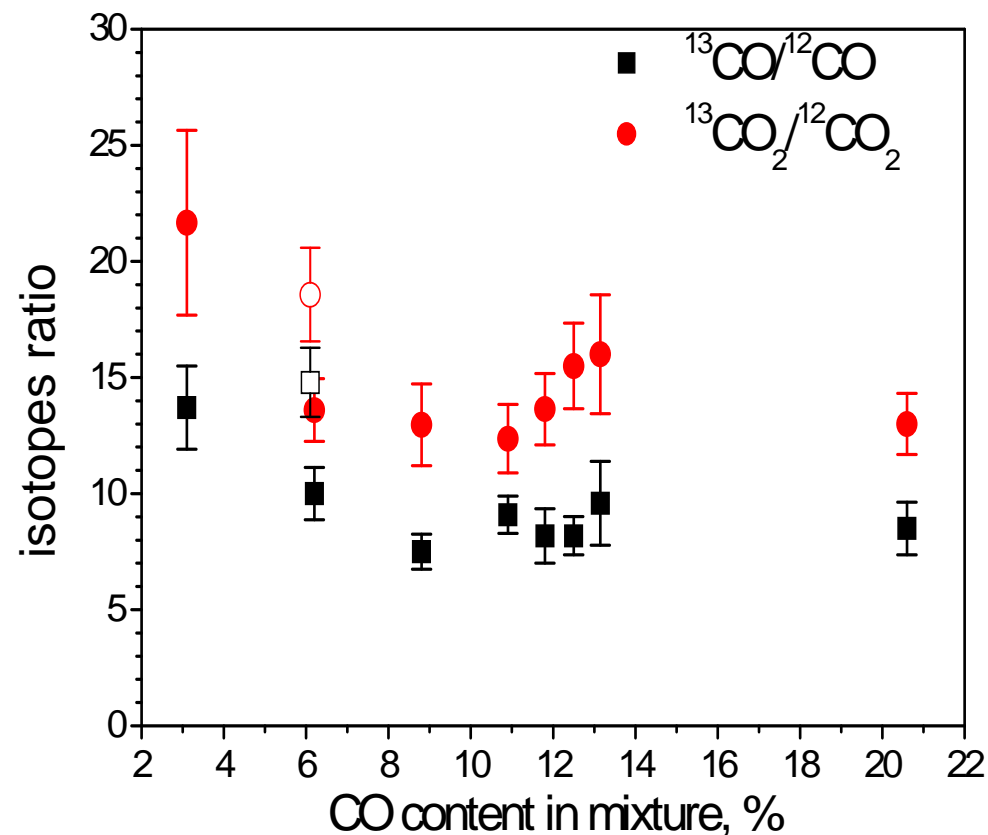
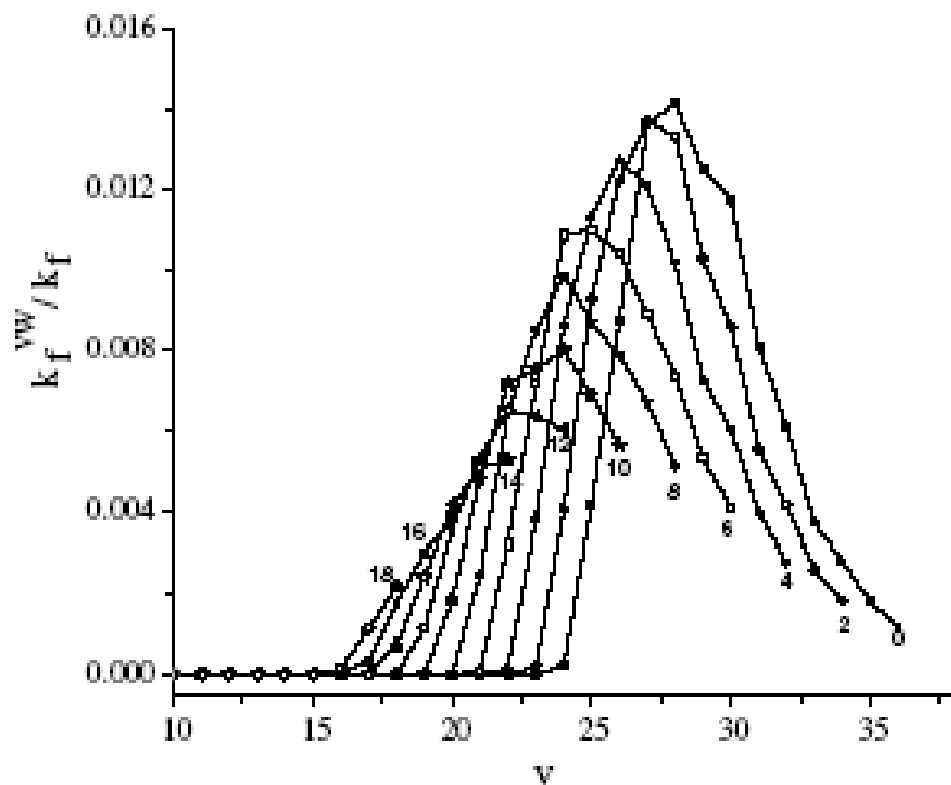




# Chemistry: Separation of Heavy Carbon Isotopes II

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## What have we learned?

- Improved methods to sustain extreme mode disequilibrium in gases: High densities, low gas kinetic temperatures, large volumes
- New data on mechanisms and rates of some critical energy transfer processes in molecular gases of aerospace interest
- Methods for selective excitation of internal energy states
- New applications: improved high power c.w. gas lasers, novel chemical syntheses, new aerodynamic control techniques

## Future directions?

- Special purpose supersonic aerodynamic testing
- Modeling code validation
- High power c.w lasers and applications: novel refrigeration?
- More chemistry: new products



# Thanks!

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## **Support:**

**USAF:**

**AFOSR Space Power & Propulsion Program**

**AFOSR Unsteady Aerodynamics and Hypersonics Program**

**AFRL Air Vehicles Directorate**

**AFRL Directed Energy Directorate**

**NASA Glenn Research Center**

**NSF**

**The Michael A. Chaszeyka Gift**

## **Collaborators:**

**Univ. of Bonn Physics Dept; Heat and Mass Transfer Institute, Belarussian  
Academy of Physics; Gas Laser Lab, Lebedev Physical Institute,  
Moscow Physical Technical Institute**

## **Kind Listeners**