

Molecular Energy Transfer Processes in Nonequilibrium Hypersonic Flows

**Igor Adamovich, Department of Mechanical and Aerospace Engineering
The Ohio State University**

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OSU NETLab (since 1986)



Faculty: Igor Adamovich and J. William Rich

Research backgrounds of students and post-docs: Mechanical Engineering, Aerospace Engineering, Electrical Engineering, Chemical Physics, Physical Chemistry, Physics

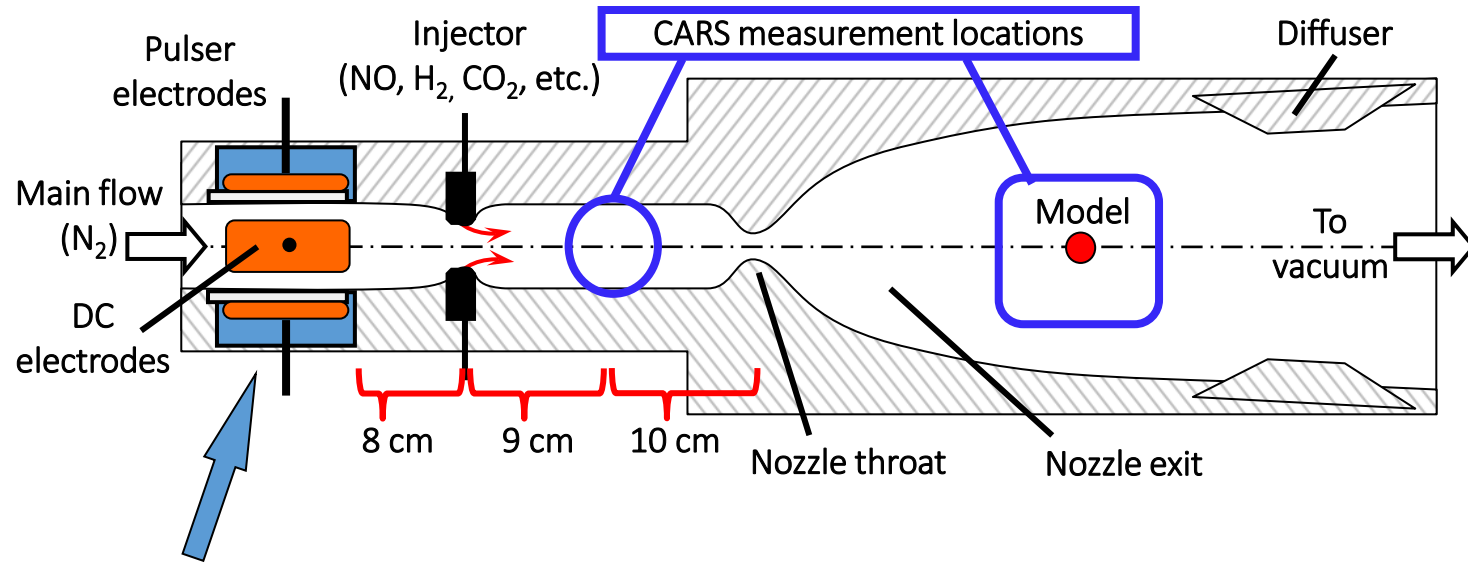
Previous state of the art

- I. Lack of laboratory scale, long run time, multiple runs per day, nonequilibrium flow facilities
 - Limits amount of flow characterization data, slows down development of diagnostics
- II. Lack of non-intrusive, high frame rate, portable diagnostics of high-speed nonequilibrium flows
 - Prevents spatially and time-resolved characterization of flow parameters, in particular state-specific measurements and kinetic model validation
- III. Lack of predictive, physics-based, state-to-state molecular energy transfer rates and kinetic models
 - Use of simplified semi-empirical models, lack of confidence in modeling predictions

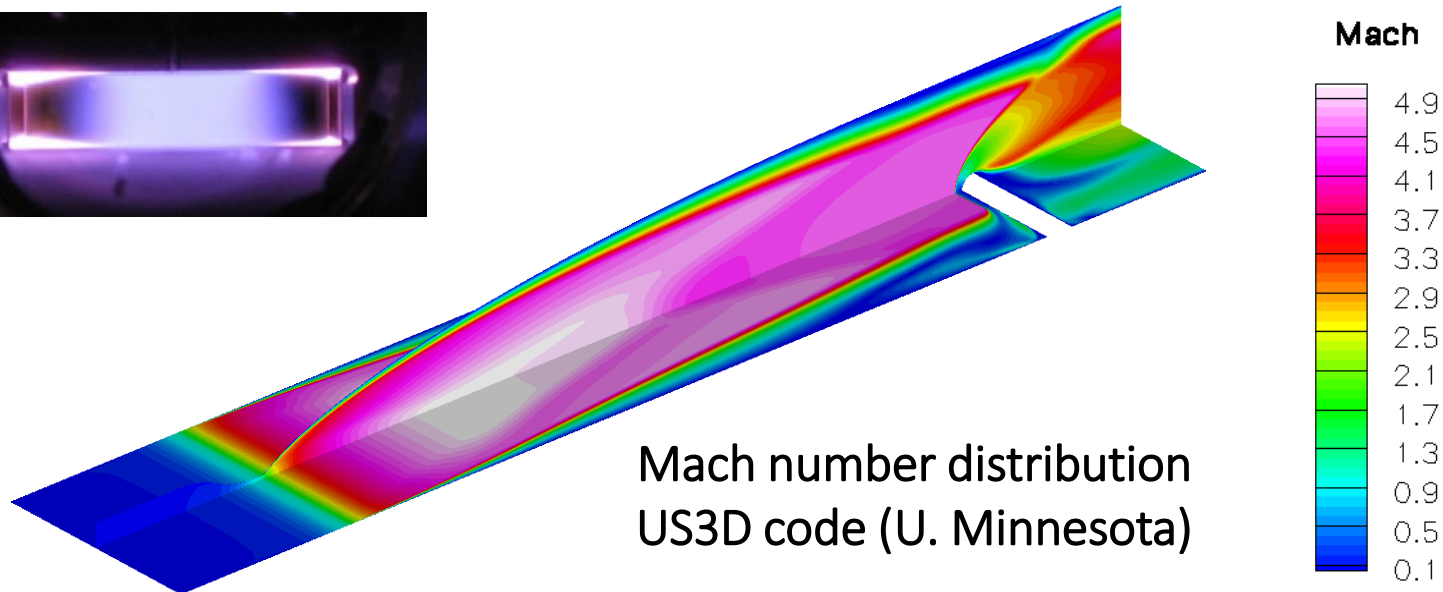
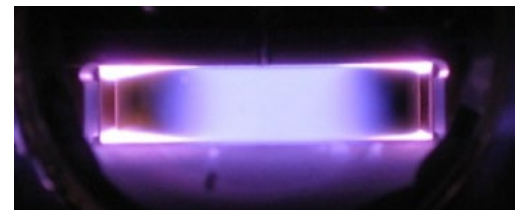
I. Development of a Nonequilibrium Flow Wind Tunnel

- Plenum pressure $P_0=0.5-1.0$ bar, steady-state run time 5-10 s
- Strong vibrational nonequilibrium generated in plenum ($T=500$ K, $T_v=2000$ K)
- Nonequilibrium flow in test section, Mach number $M=3-5$
- Used for characterization of nonequilibrium flow field by laser diagnostics
 - Ps CARS for T and T_v in plenum, Mach 5 free stream, and behind Mach 5 shock
 - 10 kHz NO PLIF for 2-dimensional temperature distribution in nonequilibrium flow behind Mach 5 shock
 - 500 kHz NO_2 / NO MTV for velocity field in flow over Mach 5 shock

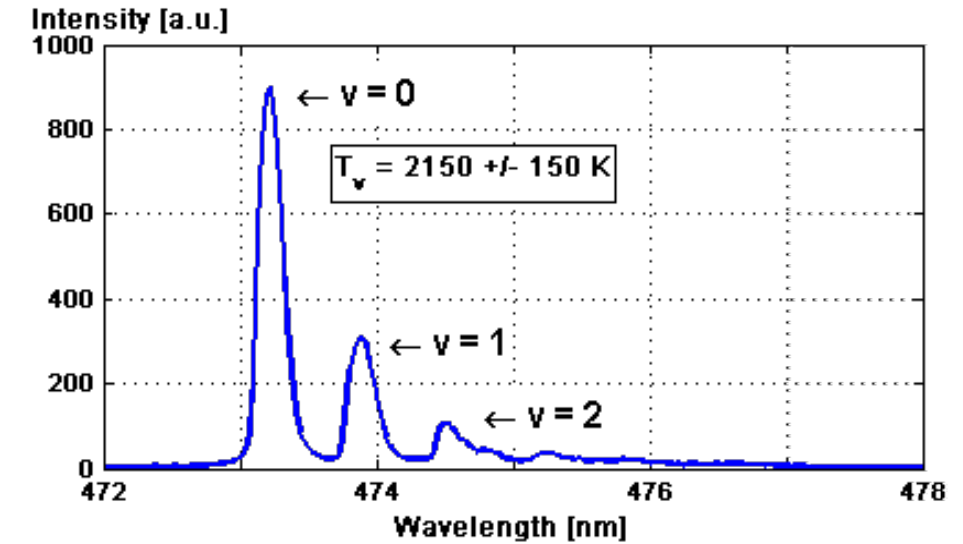
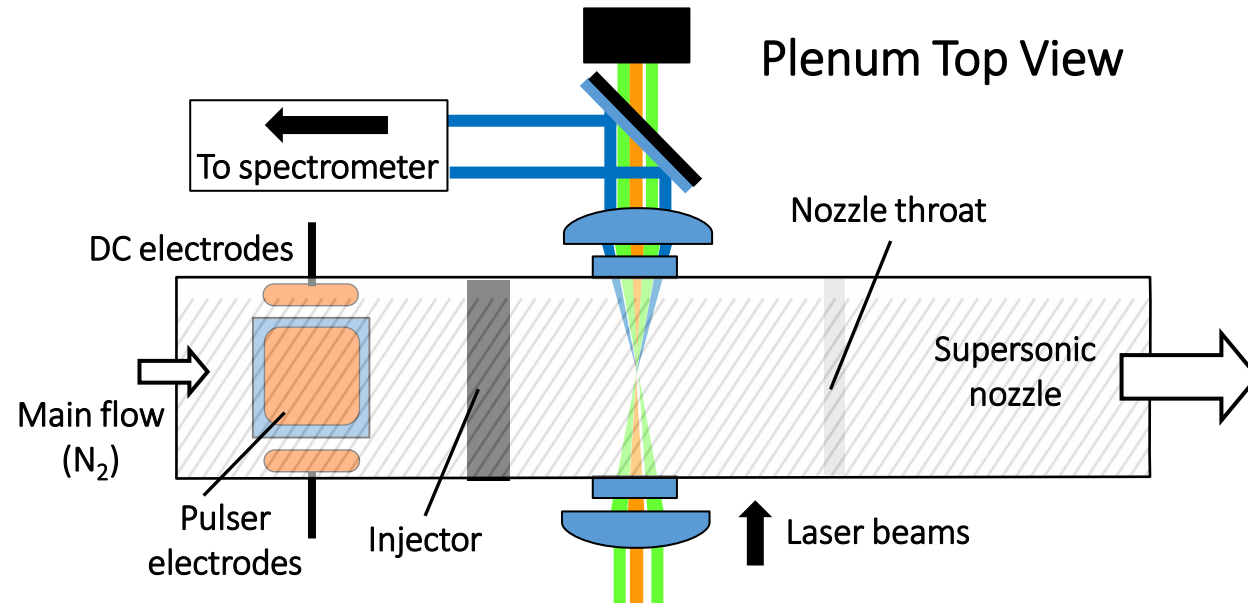
Nonequilibrium Flow Wind Tunnel Schematic



- Plenum pressure $P_0 = 0.5\text{-}1.0$ bar
- Steady-state run time 5-10 s, ~100 runs a day
- Strong vibrational nonequilibrium sustained by a diffuse electric discharge in plenum
- Interchangeable nozzle inserts, test section Mach number $M=3\text{-}5$
- Well characterized flow (US3D)
- Ample access for laser diagnostics

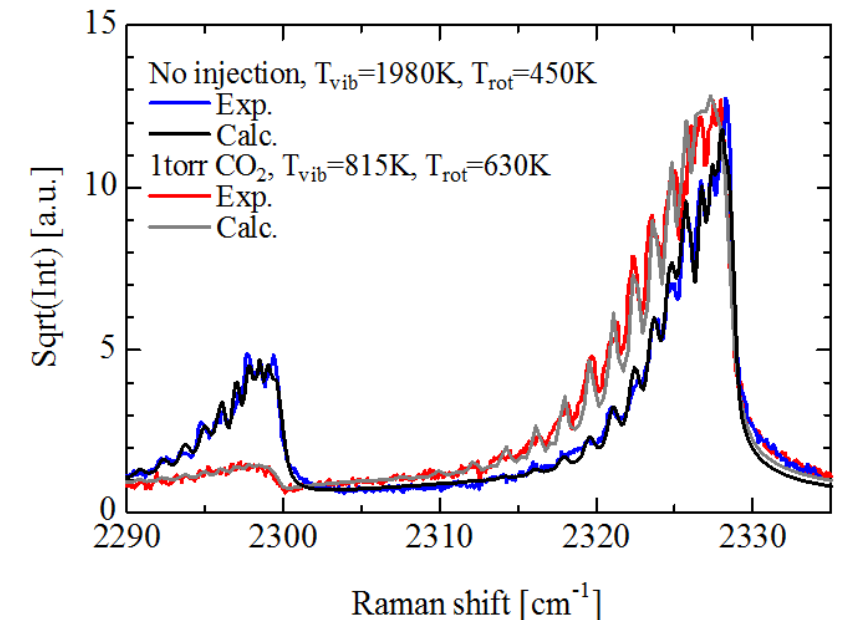


Nonequilibrium Flow Tunnel: Plenum Conditions

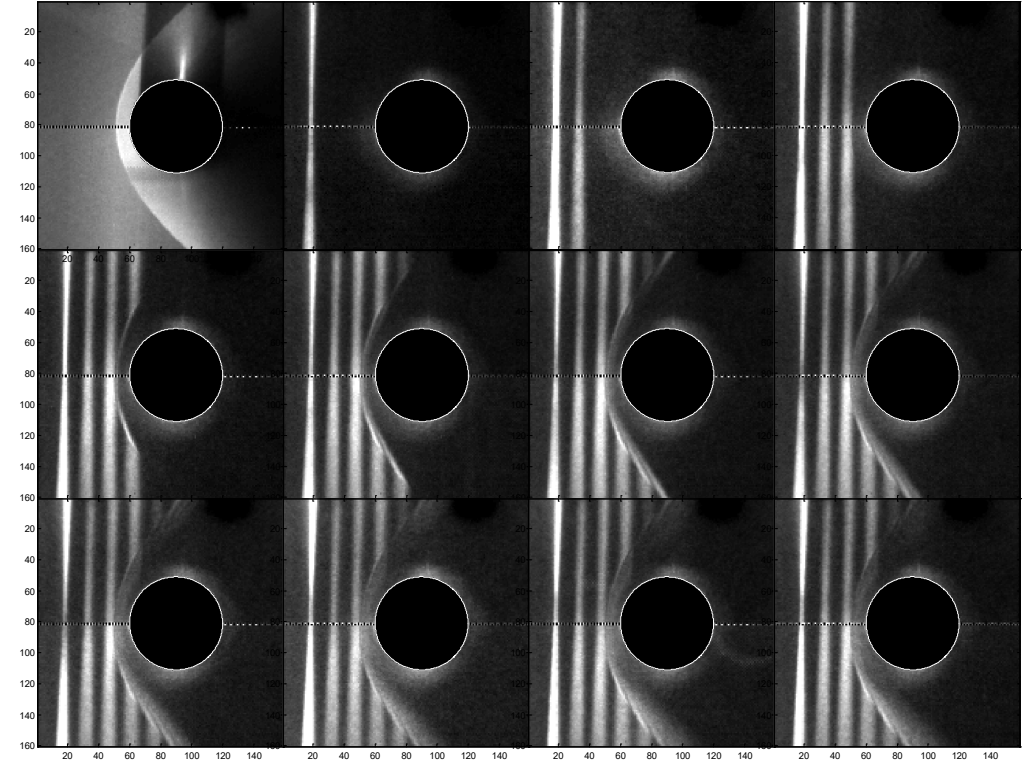
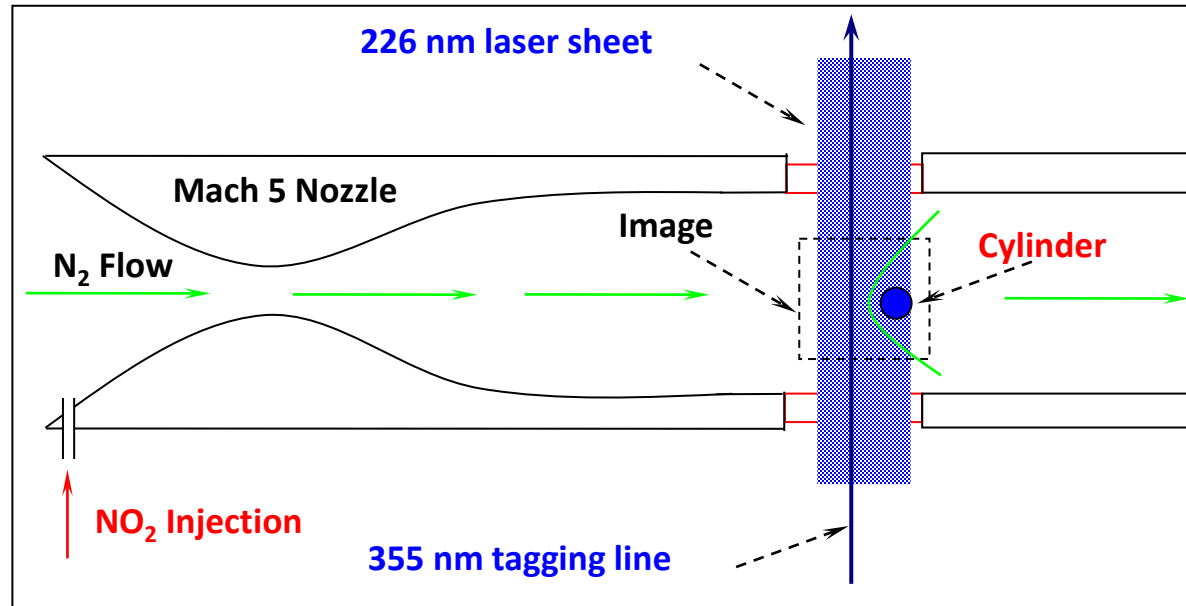


N₂ vibrational CARS spectra in plenum

- N₂(v=0-3) vibrational bands are detected
- Temperature inferred from rotational band structure
- $T_v = 2000 \text{ K}$, $T_{\text{rot}} = 450 \text{ K}$ (nitrogen);
- $T_v = 800 \text{ K}$, $T_{\text{rot}} = 600 \text{ K}$ (nitrogen with CO₂ added)
- Strong vibrational nonequilibrium at steady state
- Varied by adding relaxers such as NO, H₂, CO₂



Mach 5 Test Section: 500 kHz Flow Tagging Velocimetry*

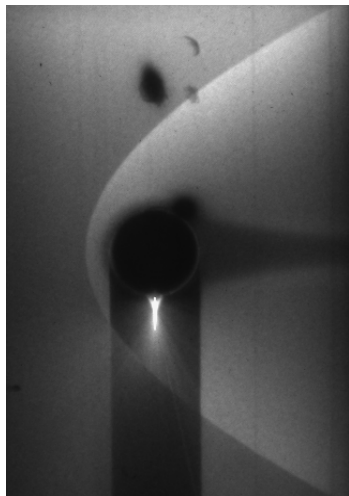
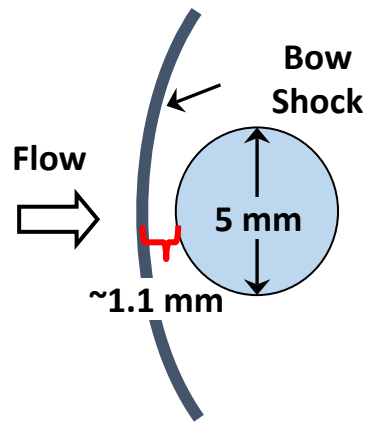


- “Tag” beam: $NO_2 + h\nu \rightarrow NO + O$ (“painting” an invisible NO line in the flow), 355 nm
- “Interrogate” sheet: NO PLIF imaging (“lighting up” the invisible lines), 226 nm
- Inferring 2-D flow velocity field in shock layer. Free stream velocity $v = 719 \pm 10$ m/s

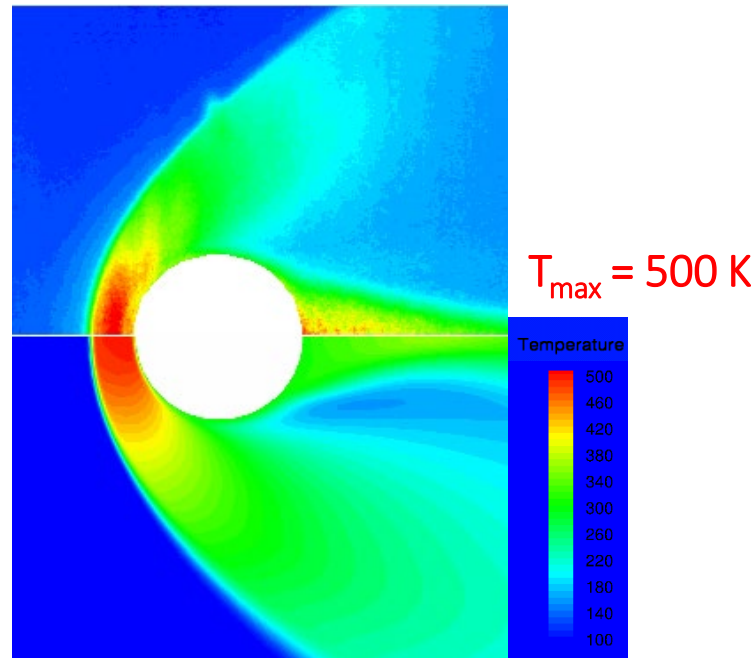
*Using OSU pulse burst laser

Nonequilibrium Mach 5 Flow Characterization: Free Stream and Shock Layer

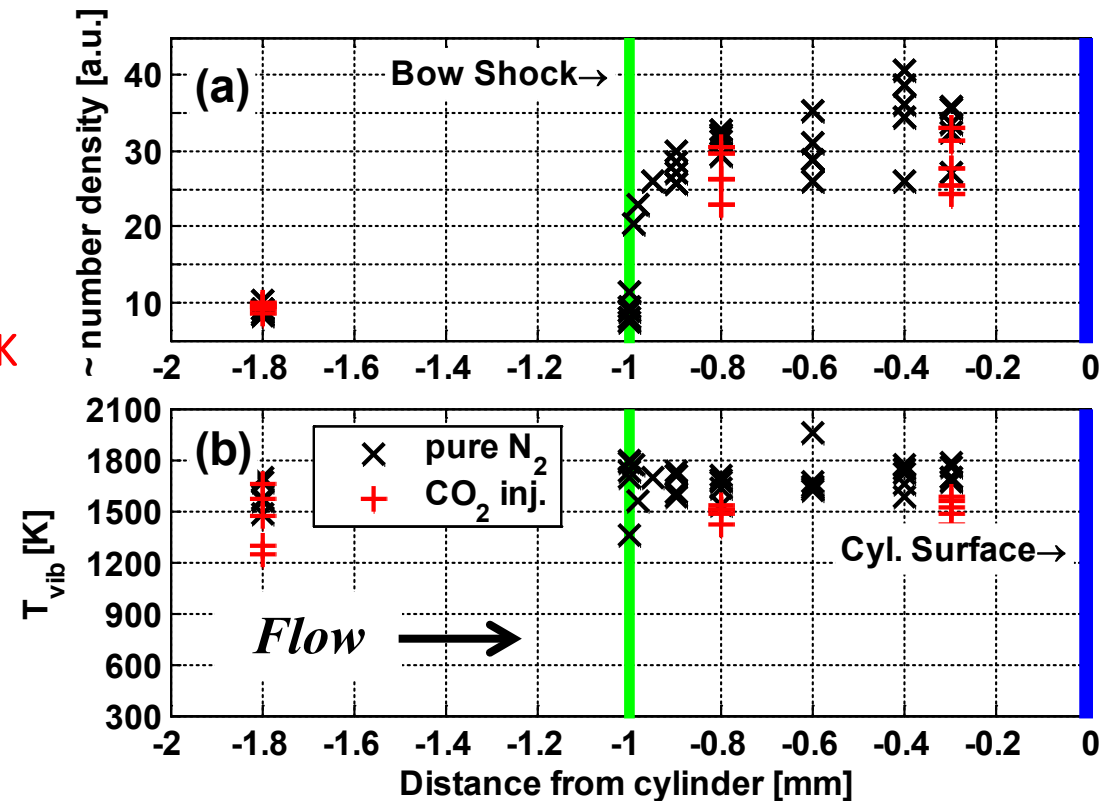
Shock in Front of a
Cylinder Model



Temperature distribution:
10 kHz NO PLIF (top), CFD (bottom)

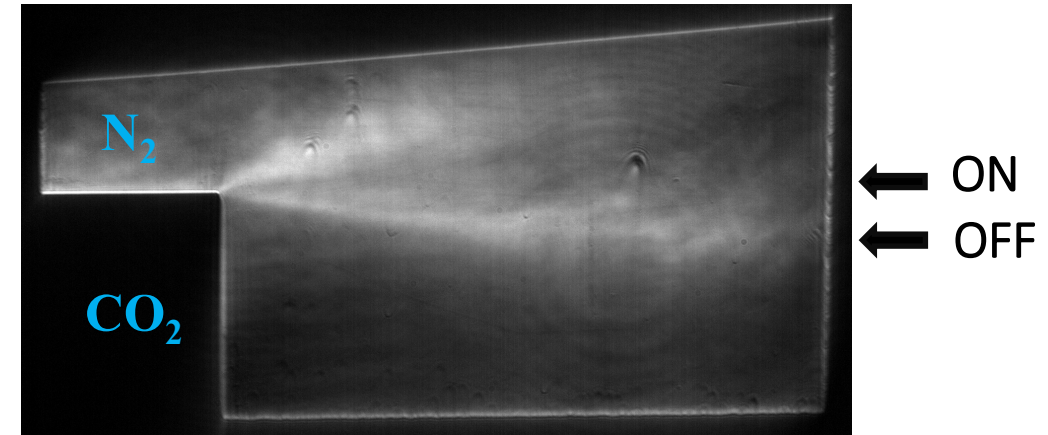
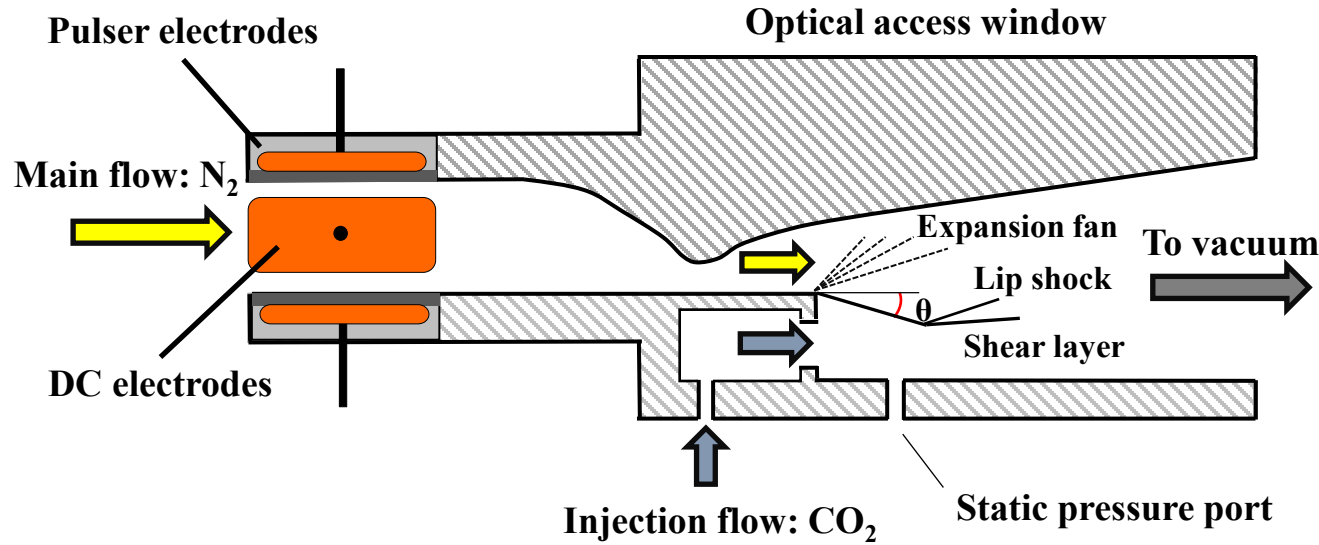


Vibrational Temperature: Ps CARS

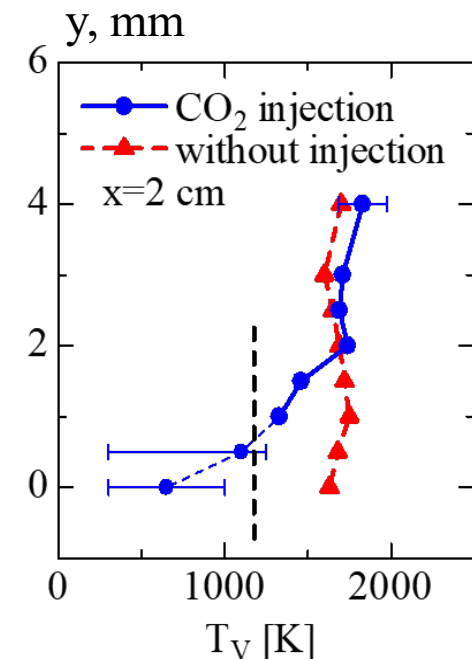


- T_{rot} , T_v distributions behind shock are measured
- “Frozen” flow behind Mach 5 bow shock: N_2 vibrational relaxation very slow

Aerodynamic Control: Vibrational Relaxation Moves Nonequilibrium Mach 3 Shear Layer



- Higher static pressure: CO_2 injection accelerates N_2 vibrational relaxation
- This results in gas temperature and pressure rise, pushing shear layer up
- 2-D N_2 vibrational temperature distribution measured in shear layer
- Effect observed only when N_2 is vibrationally excited
- At $T > T_v$, effect would be reversed (vibrational relaxation would reduce T)



Impact

- Robust, laboratory scale experimental platform for detailed studies of nonequilibrium hypersonic flows
- Essential for development and testing of laser diagnostics
- Straightforward generation of nonequilibrium high-pressure flows
- Detailed characterization of nonequilibrium flow in plenum, free stream, and Mach 5 shock layer
- Effect of accelerated vibrational relaxation on Mach 3 shear layer is detected and quantified
- Effect may be observed behind oblique shocks, in base flows in hypersonic flight

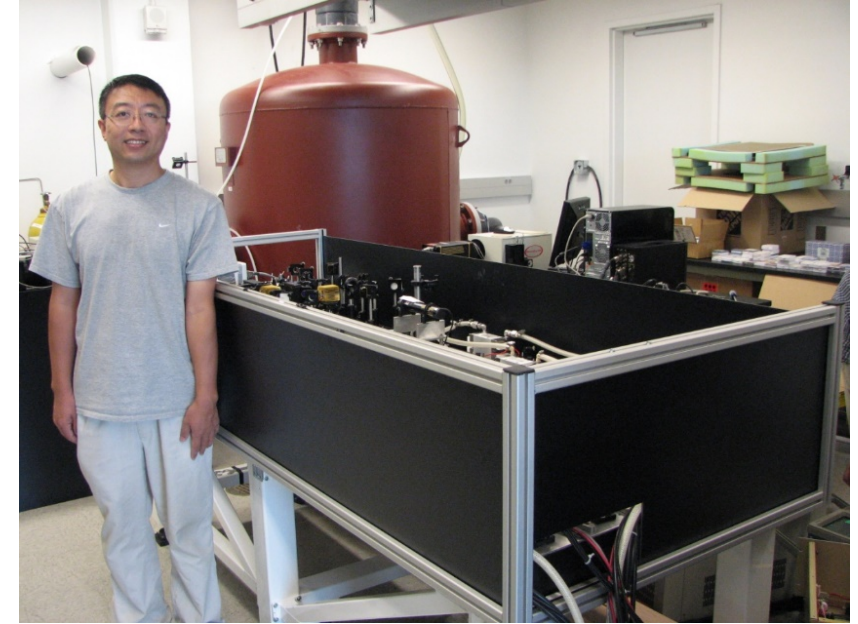
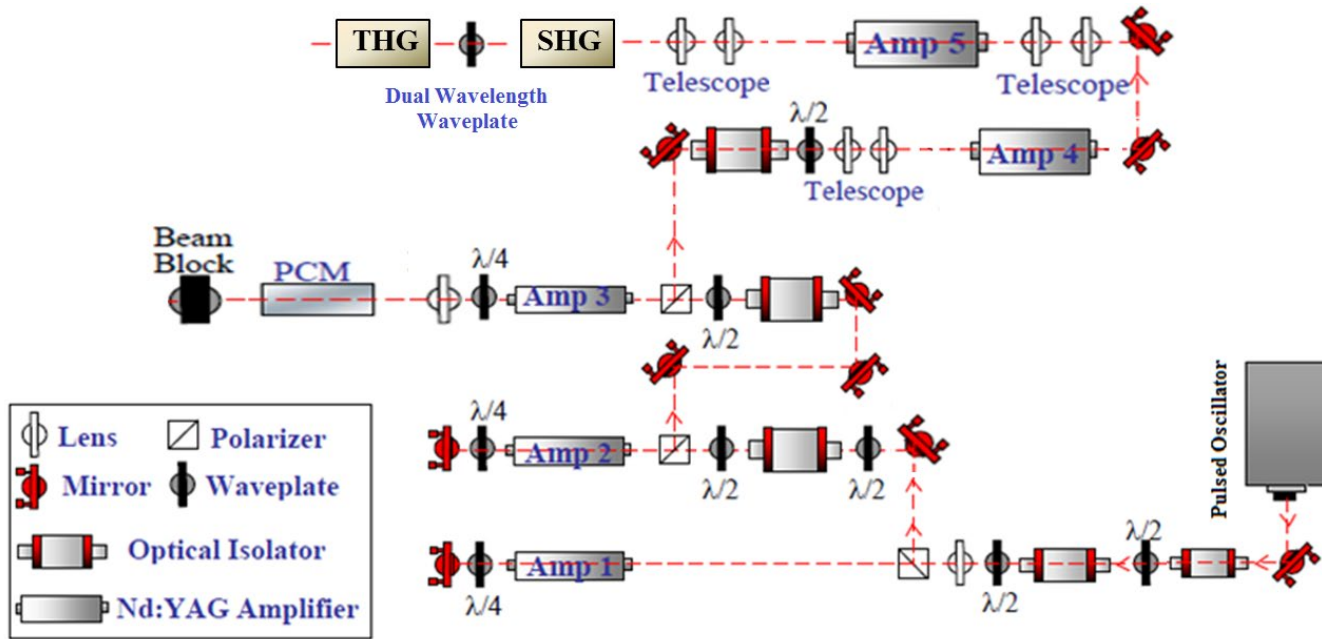
Challenges

- Effect of vibrational relaxation on high-speed flow field needs to be quantified at well characterized flow conditions, compared with nonequilibrium flow code predictions
- Time-accurate effect of vibrational relaxation on shock stand-off distance: high frame rate NO PLIF
- Development of high frame rate CARS (CARS data so far are obtained at 10 Hz)
- 10 Hz CARS limitation: although 10-100 laser shots used for 1-10 s wind tunnel run time, only one laser shot per run is used at short run time facilities
- Development of Cavity Ring Down Spectroscopy diagnostic, to measure “dark” states (non-radiating metastable species) in the flow, $N_2(A^3\Sigma)$ and $O_2(a^1\Delta)$
- These states are critical for quantifying UV emission behind the shock, O_2 dissociation kinetics

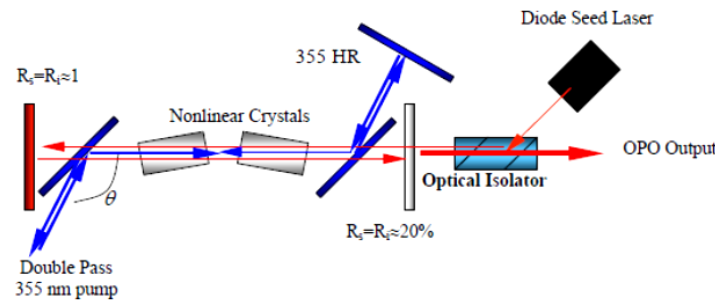
II. Development of Laser Diagnostics of High-Speed Flows

- OSU pulse burst laser / high frame rate flow imaging system
 - Custom built Nd:YAG laser outputs “bursts” of 10-30 high energy, ns duration pulses at rep rate of up to 1 MHz (1 μ s apart)
 - Pulse energy \sim 100 mJ (@1064 nm) at 1 MHz to \sim 500 mJ at 10 kHz
 - Tunable UV output generated by Optical Parametric Oscillator (OPO), in combination with sum frequency mixing
 - Planar Laser Induced Fluorescence (PLIF) imaging captured with high frame rate cameras
 - At OSU, used for 10 kHz NO PLIF and 500 kHz Molecular Tagging Velocimetry
- The laser is portable, traveled to take data at hypersonic flow facilities at NASA LaRC and CUBRC

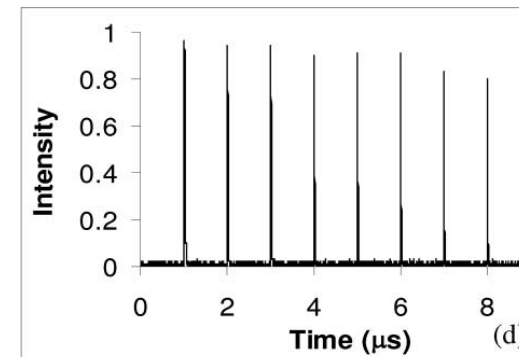
Pulse Burst Laser / Flow Imaging System



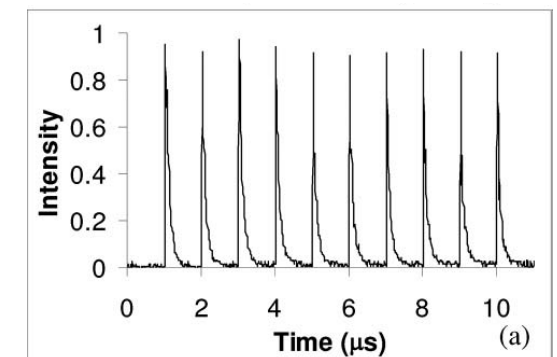
Typical 1 MHz laser pulse trains



Low energy, 1064 nm fiber laser pulses undergo 5 stages of amplification, frequency tripled to pump narrow linewidth OPO

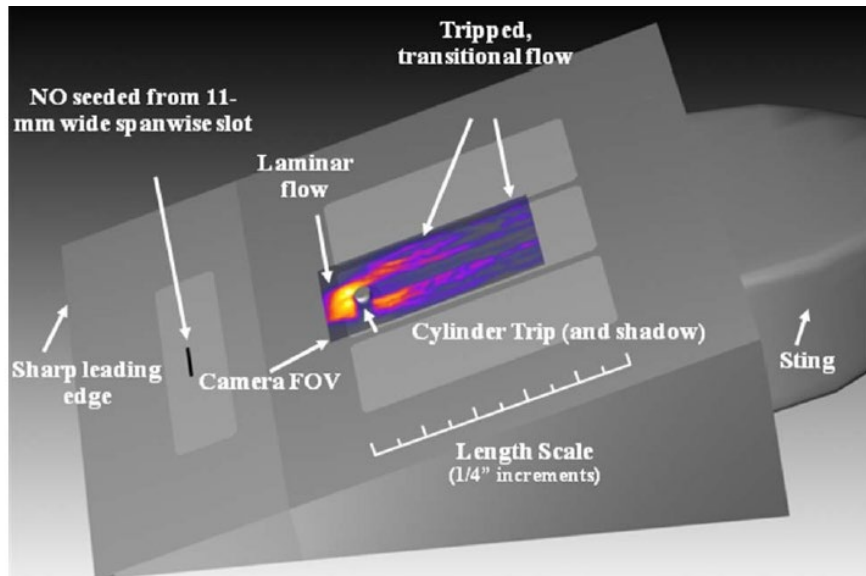
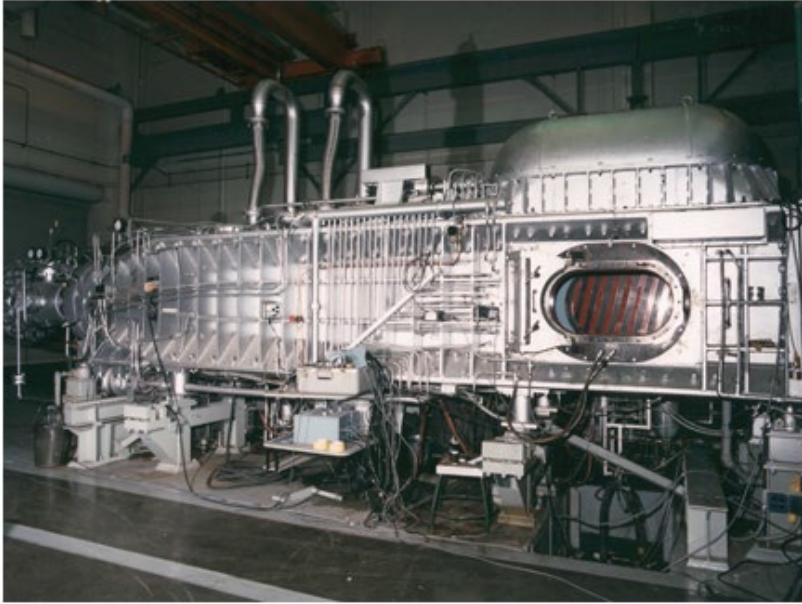


1064 nm (100 mJ/pulse)

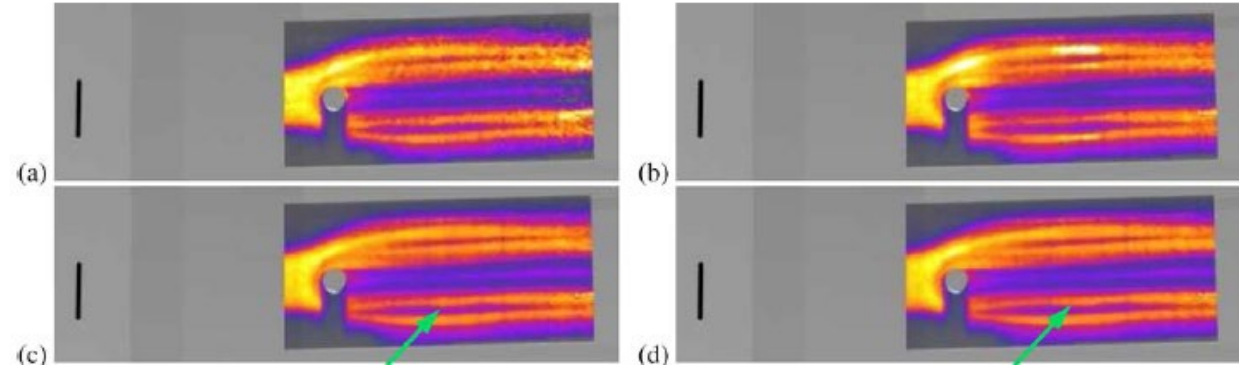


226 nm for NO LIF (0.4 mJ/pulse)

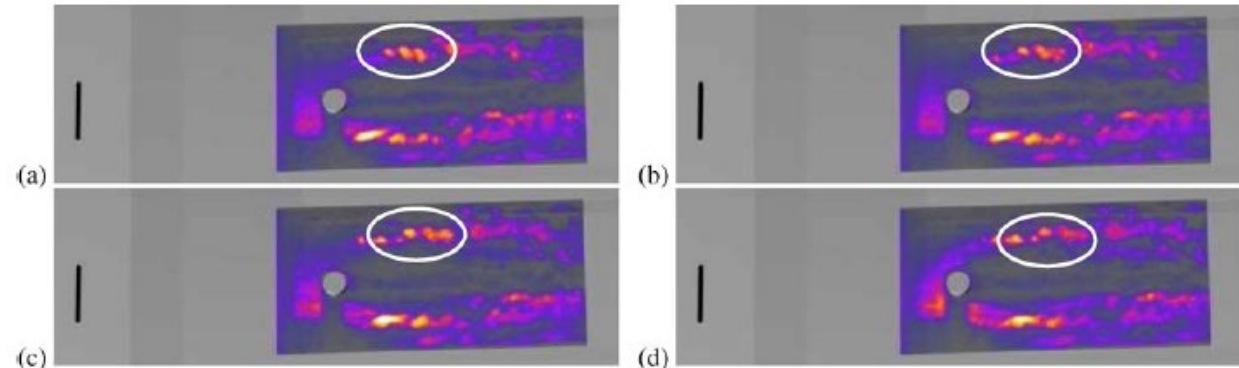
NO PLIF at NASA LaRC Mach 10 Wind Tunnel



$$Re = 1.7 \cdot 10^6 / m$$

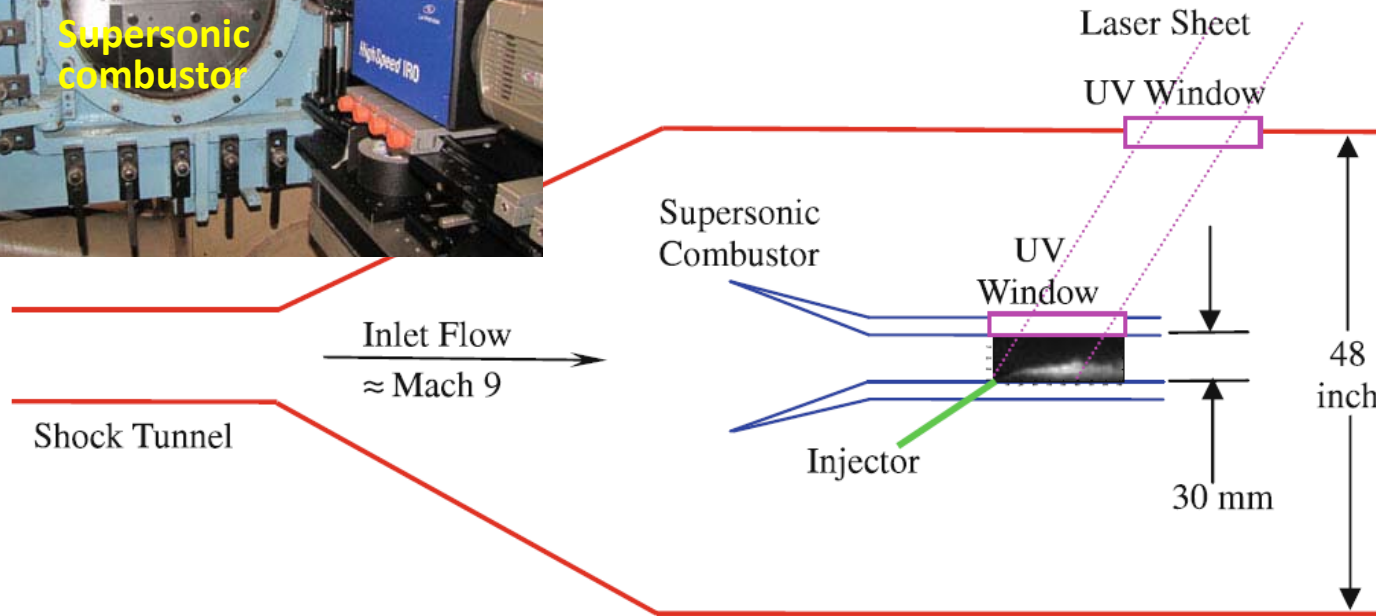


$$Re = 6.2 \cdot 10^6 / m$$



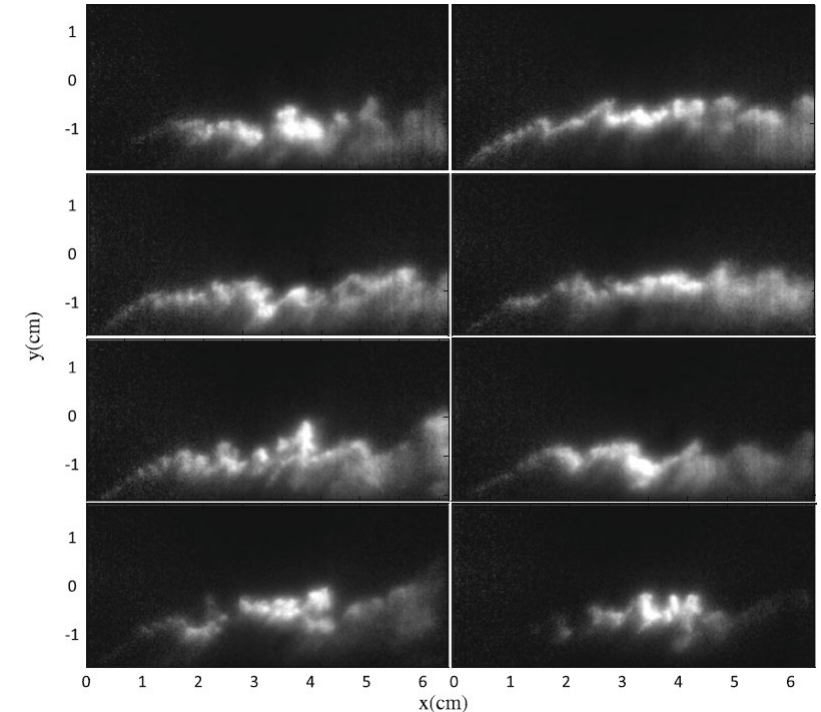
- Multiple sets of 500 kHz NO PLIF imaging of a Mach 10 boundary layer after a cylindrical trip, $Re = 1.7 - 6.2$ million / m
- Flow ranges from laminar to highly transitional, with instabilities and corkscrew vortices identified

NO PLIF at CUBRC 48" Shock Tunnel

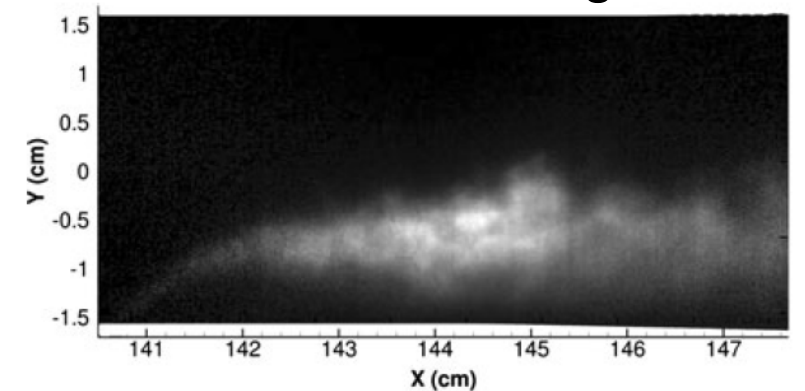


- Free stream flow: nitrogen, Mach 9, run time 10 ms
- 10 kHz NO PLIF images of NO-seeded He jet injected into a model supersonic combustor
- Data quantify jet penetration depth and mixing with the main flow

Single frames



10-frame average



Impact

- OSU pulse-burst laser: portable diagnostics for high frame rate characterization of nonequilibrium flows (~10 data sets per ~1 ms run)
- Diagnostics development, “shake-down”, and testing made possible by using the OSU laboratory scale nonequilibrium flow wind tunnel
- 500 kHz imaging of Mach 10 laminar and transitional boundary layer at NASA LaRC
- 10 kHz imaging of injection into a model supersonic flow combustor at CUBRC
- 500 kHz Mach 5 flow velocimetry at OSU

Challenges

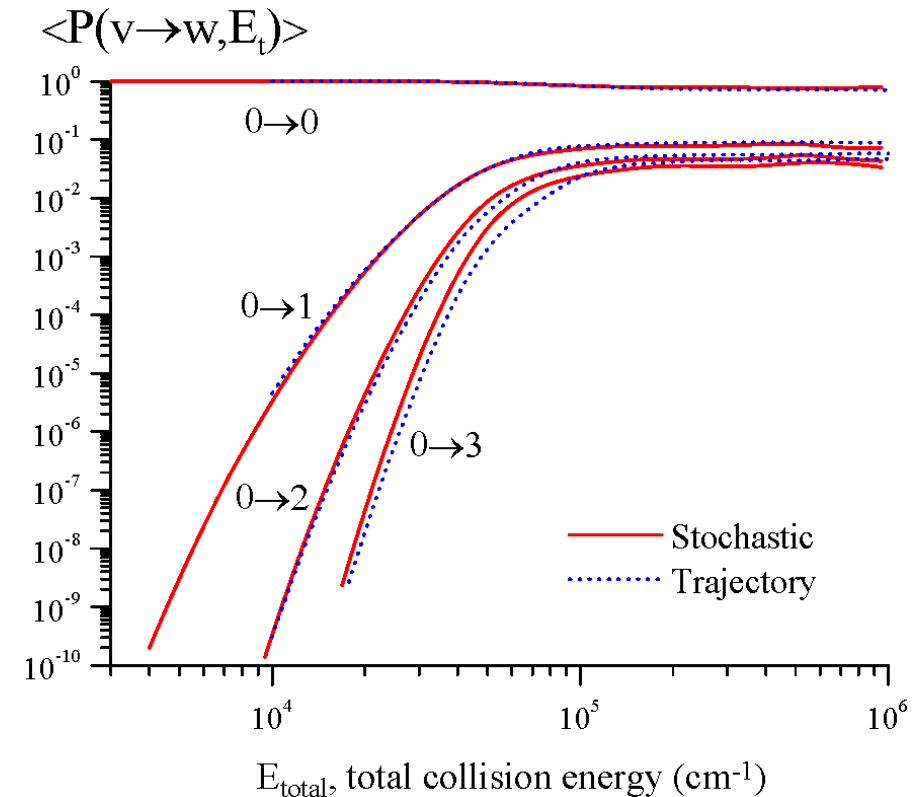
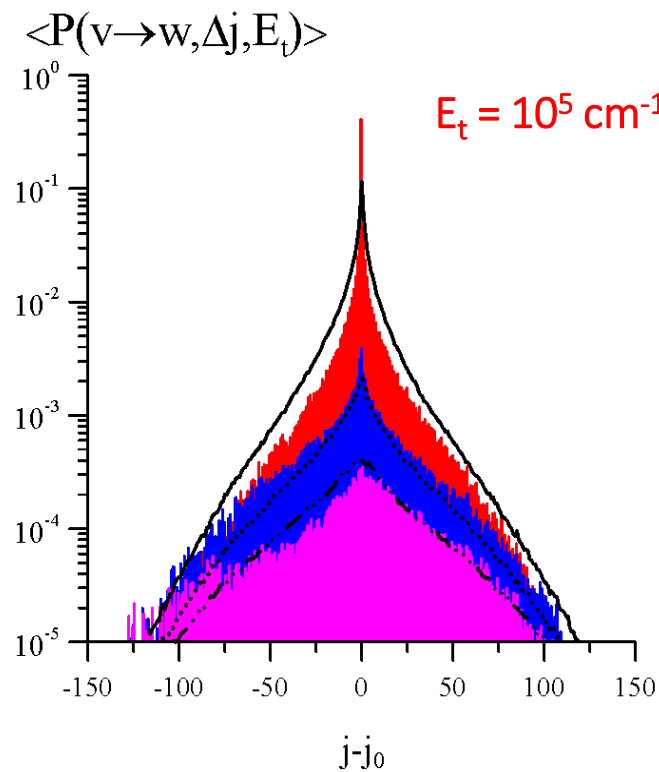
- Development of portable, high frame rate CARS diagnostics, using the OSU pulse burst laser combined with a broadband OPO
- All previous CARS data are obtained at 10 Hz, such that only a single laser shot per run can be used at short run time test facilities.
- Use of high frame rate CARS will increase rate of data acquisition by an order of magnitude (~ 10 - 20 data sets for each ~ 1 ms run)
- Major technical challenge: monitoring and quantifying shot-to-shot variation of the Stokes beam spectral profile. This is critical for quantitative high frame rate CARS measurements.
- Operating the OSU nonequilibrium wind tunnel and the existing 10 Hz CARS setup is essential for development of this diagnostic.

III. Development of Molecular Energy Transfer / Nonequilibrium Air Chemistry Models

- Forced Harmonic Oscillator – Coupled Rotation model: physics-based, state-specific, close-coupled vibrational-rotational-translational (V-R-T) rates
- Coupled R-T / V-T energy transfer is critical near molecular dissociation limit
- 3-D molecular collisions
- Coupling among multiple vibrational levels, coupling between vibrational and rotational energy transfer
- Multi-quantum V-R-T rates predicted at high collision energies (temperatures)
- Good agreement with computer trajectory calculations for accurate potential energy surfaces
- Coordinated model development, experimental work, and validation in the same laboratory

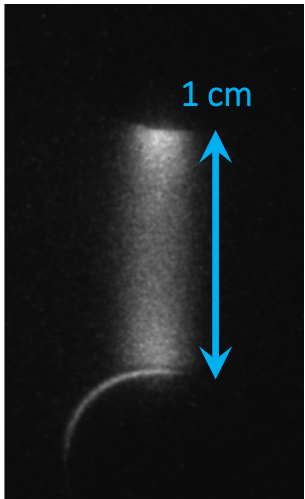
Comparison with Trajectory Calculations:

$P_{VRT}(v, j_0 \rightarrow w, j)$ and $P_{VT}(v \rightarrow w)$ for Nitrogen

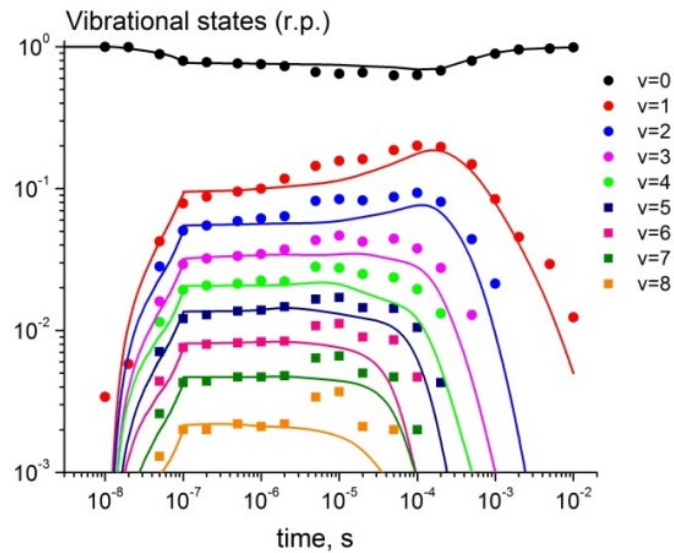


- Good agreement with 3-D computer trajectory calculations for an accurate potential energy surface
- Analytic rate expressions: straightforward incorporation into existing nonequilibrium NS and DSMC codes
- Physics-based, predictive analysis of energy transfer and nonequilibrium chemistry behind strong shocks

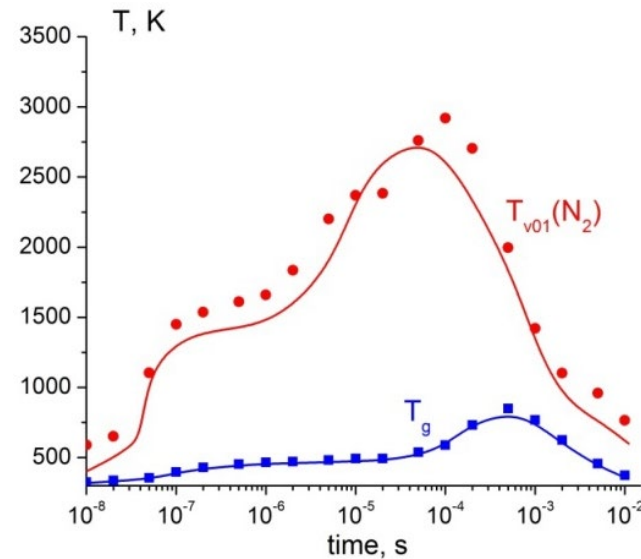
Comparison with State-Specific Vibrational Energy Transfer Measurements in Air Plasma



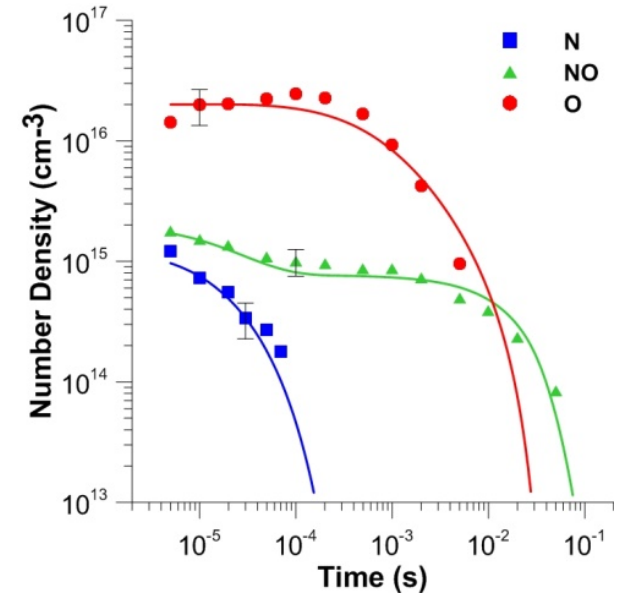
NO PLIF image



N₂(v=0-8) populations



Vibrational temperature
Gas temperature



[N], [O], [NO]

- Pulsed air excitation in a diffuse filament, ~100 ns pulse discharge, P=100 Torr
- Experimental data: ps CARS (N₂ vibrational level populations), ns LIF and TALIF (NO, O, N number densities)
- Good agreement between data and modeling predictions for time-resolved N₂(v=0-8) vibrational level populations, vibrational temperature, gas temperature, and [N], [O], [NO]

Impact

- Straightforward incorporation of the model into nonequilibrium flow codes (Candler 1997; Boyd and Josyula 2011; Levin 2012; Schwartzentruber 2014; Gimelshein and Wysong 2018)
- Physics-based analysis of energy transfer and nonequilibrium chemistry at strongly nonequilibrium conditions
- Straightforward analysis of the molecular Potential Energy Surface (PES) effect on the energy transfer rates
- Complementing higher-fidelity, accurate PES, adiabatic / nonadiabatic trajectory calculations

Challenges

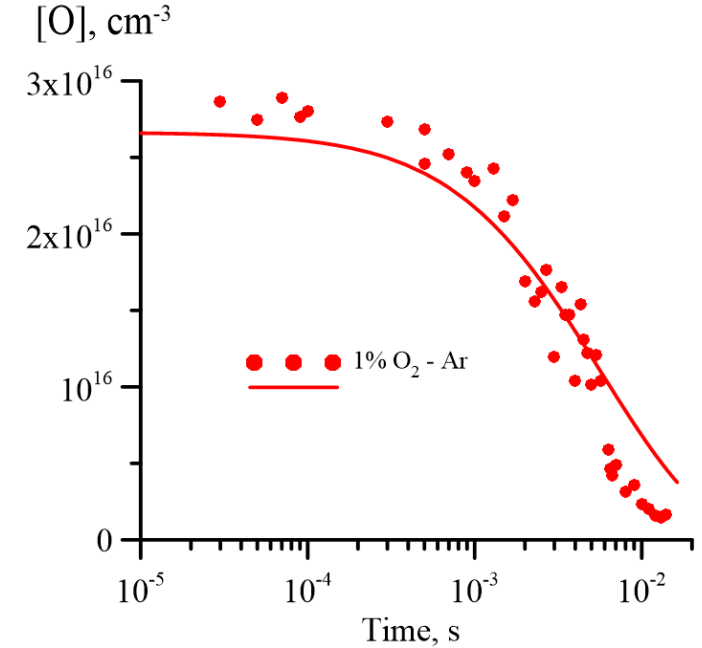
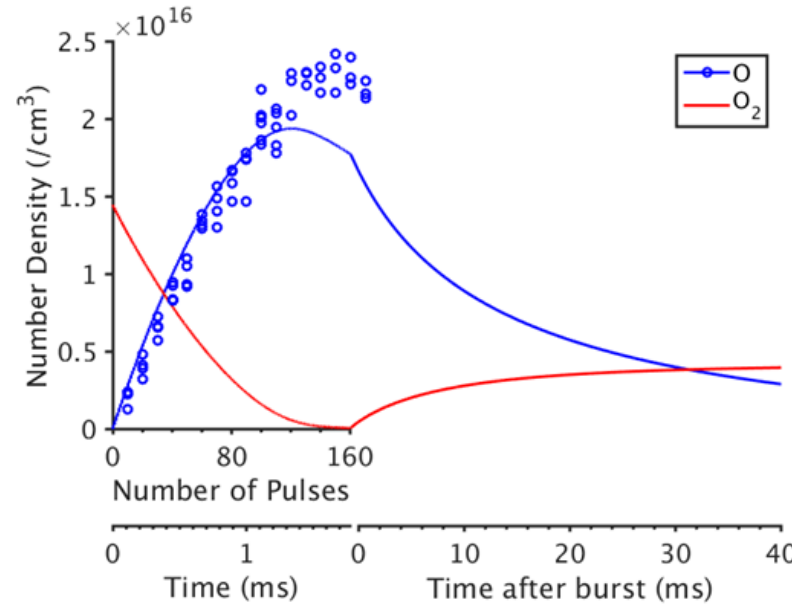
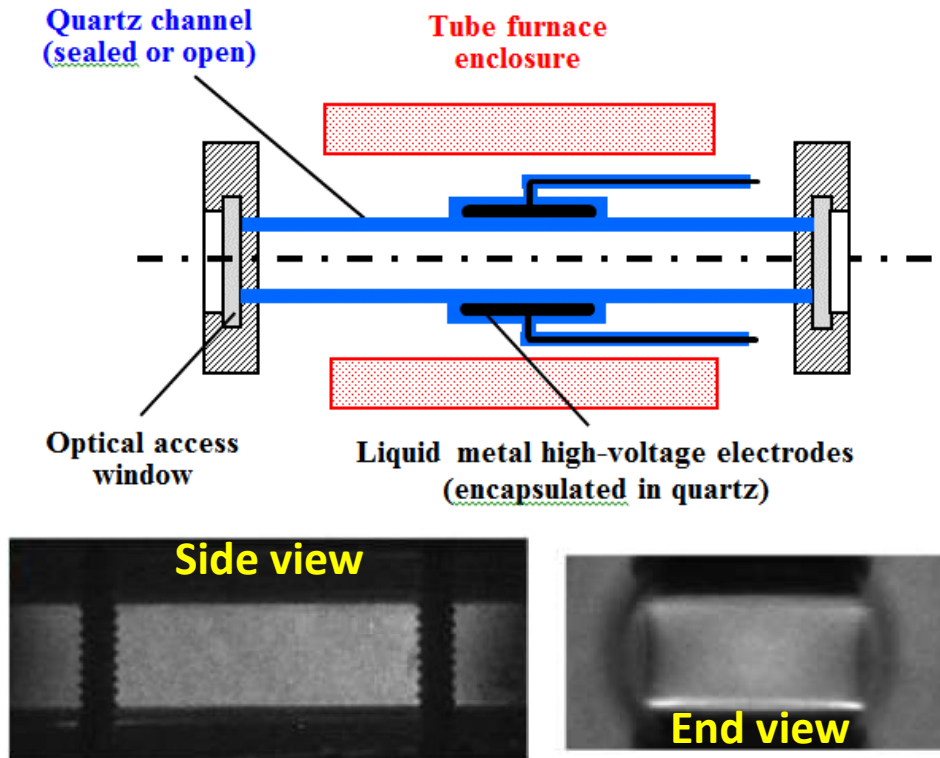
- High-fidelity, accurate PES, state-specific vibrational energy transfer and dissociation models have been developed recently (U. Minnesota, U. Michigan)
- Model validation data, specifically for state-specific dissociation rates, are scarce to nonexistent
- Need relevant experimental data obtained at well characterized conditions

Ongoing and Future Research - I

- What are the state-specific rates of O_2 dissociation behind strong shocks (above $M \sim 6$)? O_2 dissociation drives high-temperature nonequilibrium air chemistry, UV and IR emission behind the shock
- How can recent high-fidelity predictions of these rates (Boyd et al. 2015) be validated? State-specific measurements of $O_2(v)$ and ground state O atoms in shock tubes are extremely challenging
- Approach: simultaneous time-resolved measurements of O atoms and $O_2(v)$ in recombining O atom - Ar buffer mixture, at well characterized conditions, obtaining dissociation rates from detailed balance:



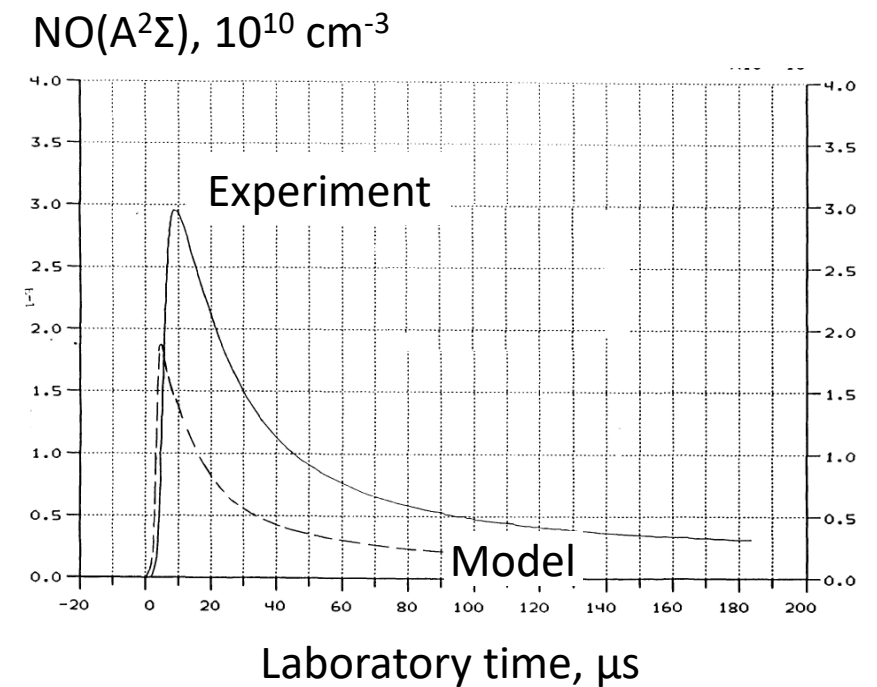
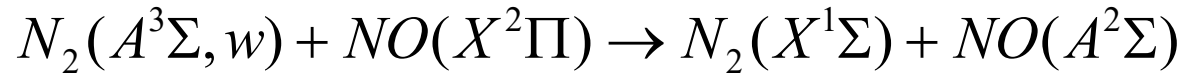
State-Specific Measurements of O atoms and $O_2(v)$ During Recombination



- O_2 -Ar excitation by a uniform ns pulse discharge burst (~ 100 pulses at 100 kHz), at $P=0.5-1.0$ atm, $T_0=500$ K
- Complete dissociation of O_2 by electron impact, partial pressure of O atoms ~ 1 Torr
- Time-resolved measurements of O atoms (ps TALIF) and $O_2(v)$ (ps LIF) during O + O recombination
- Comparison with modeling predictions, validating state-of-the-art dissociation model (U. Michigan)

Ongoing and Future Research - II

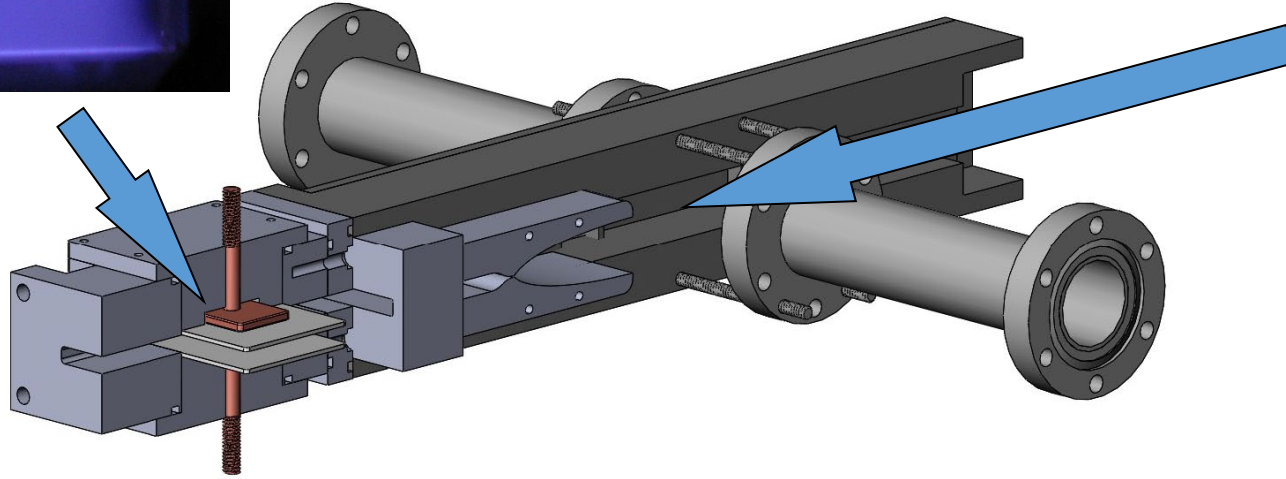
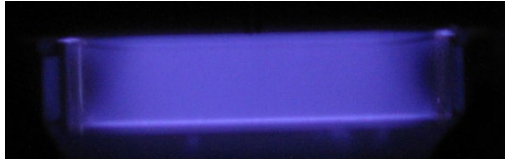
- What is the effect of metastable (“dark”) molecular states on UV radiation from strong shocks (M=8-11)?
- Modeling predictions: NO UV radiation (γ bands) is due to energy transfer from metastable excited nitrogen, $N_2(A^3\Sigma)$ (Wurster 1991, Treanor 1993)



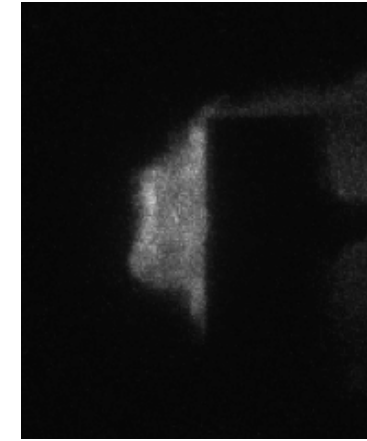
- Approach: generate $N_2(A^3\Sigma)$ in an electric discharge in wind tunnel plenum

NO UV emission behind a normal shock
in air ($u_s=3.86 \text{ km/s}$)

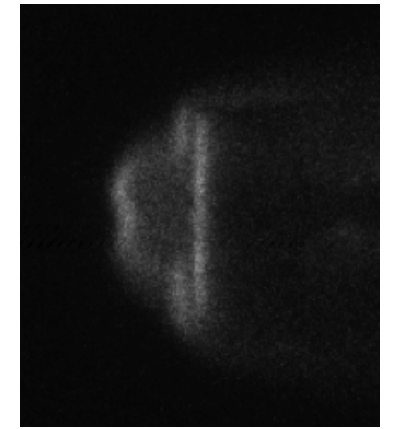
$N_2(A^3\Sigma)$ and NO measurements in Mach 5 Flow



NO injected upstream
from a cylinder model



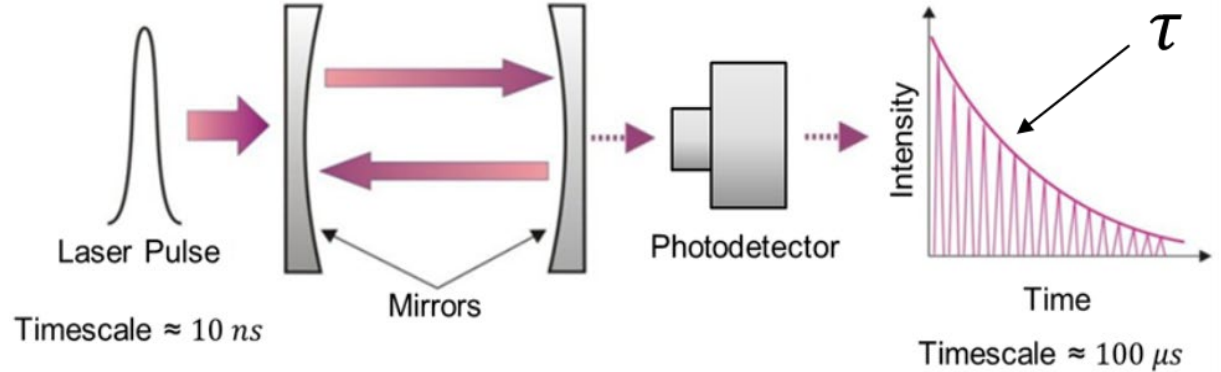
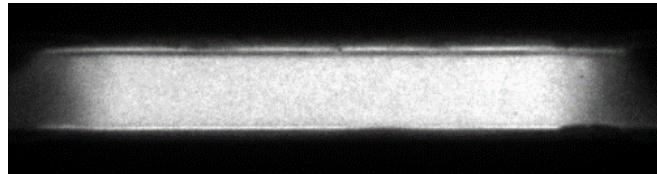
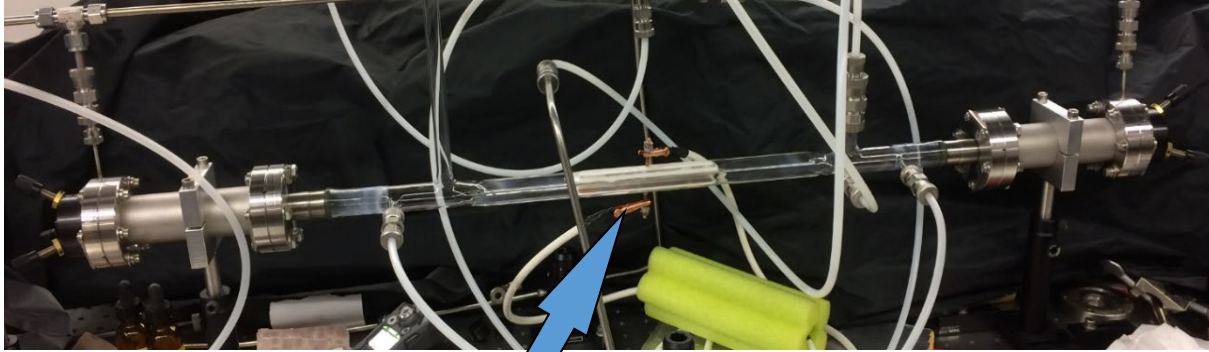
NO PLIF



NO(A \rightarrow X) emission

- Generate $N_2(A^3\Sigma)$ in uniform ns pulse discharge burst in wind tunnel plenum
- Seed the flow with NO (in plenum or in supersonic flow section)
- Measure $N_2(A)$ and NO(X) in a Mach 5 flow: CRDS, NO PLIF
- Image NO(A \rightarrow X) UV emission (γ bands), quantify energy transfer from $N_2(A)$ to NO(A)
- Generate data at well characterized flow conditions, compare with NO UV emission predictions by nonequilibrium flow codes

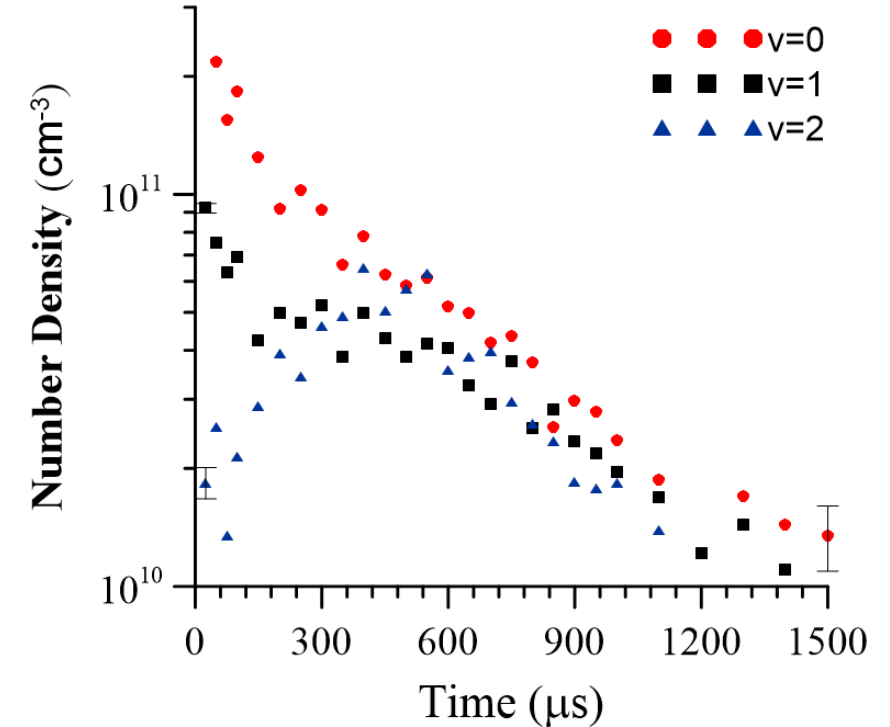
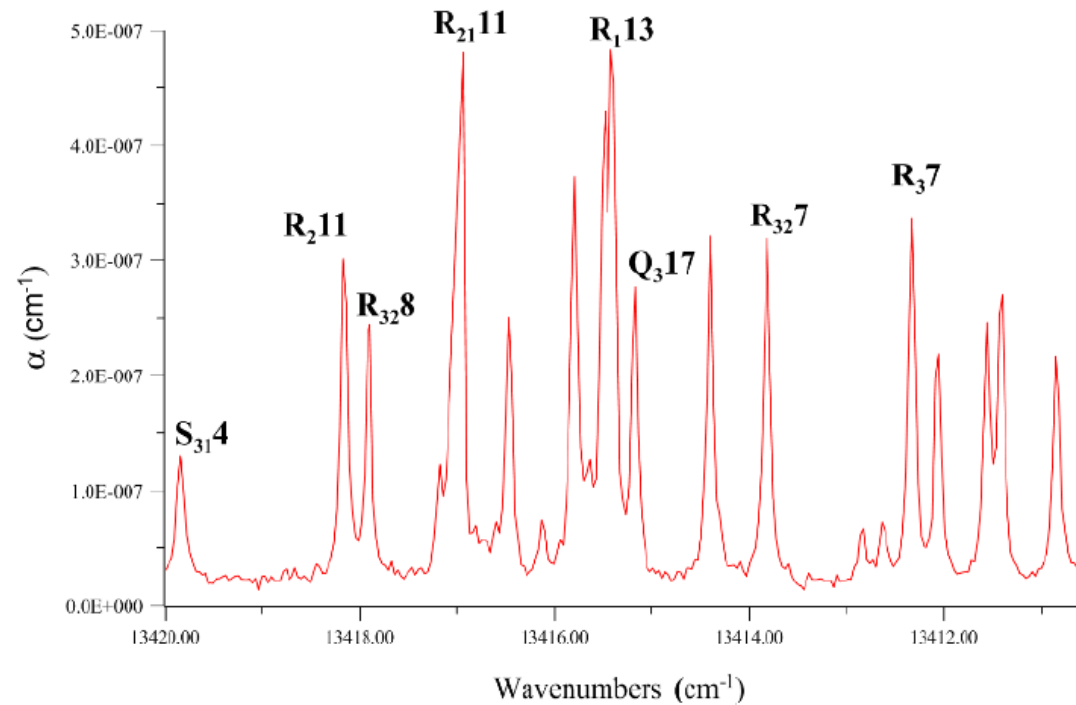
Cavity Ring Down Spectroscopy Diagnostics for $N_2(A^3\Sigma)$ Measurements in the Flow



$$\frac{I}{I_0} = \exp\left(-\frac{t}{\tau}\right) \quad [N_2(A^3\Sigma)] = \frac{1}{c\sigma_{abs}} \left(\frac{1}{\tau} - \frac{1}{\tau_{empty}} \right)$$

- $N_2(A^3\Sigma)$ generated in uniform nitrogen plasma in CRDS cavity by a ns pulse discharge burst
- Absorption of a laser pulse is measured in the cavity with 99.99% reflectivity mirrors
- Absorption path ~ 2 km, very high sensitivity $N_2(A, v=0-2)$ measurements after the burst

Results So Far: $\text{N}_2(\text{A}^3\Sigma, v=0-2)$ in Nitrogen Plasma*



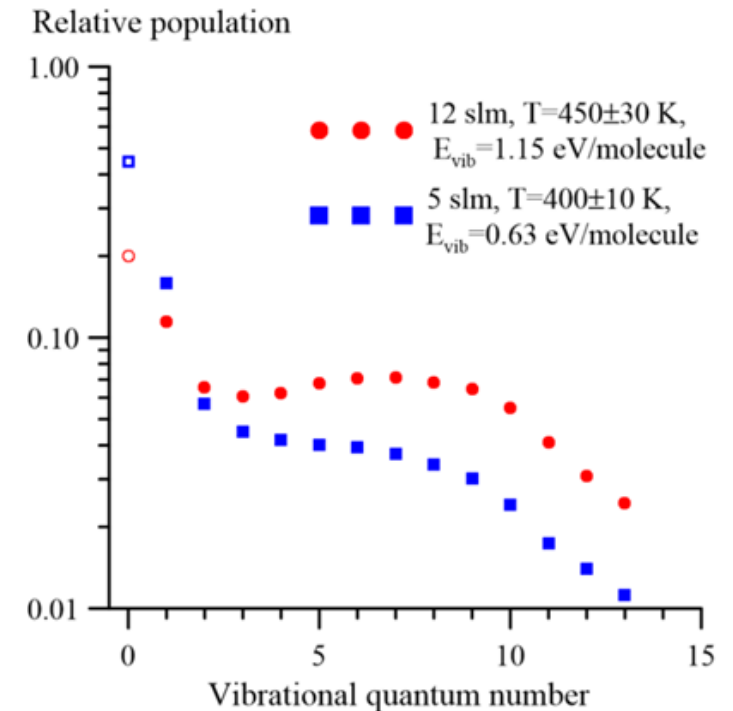
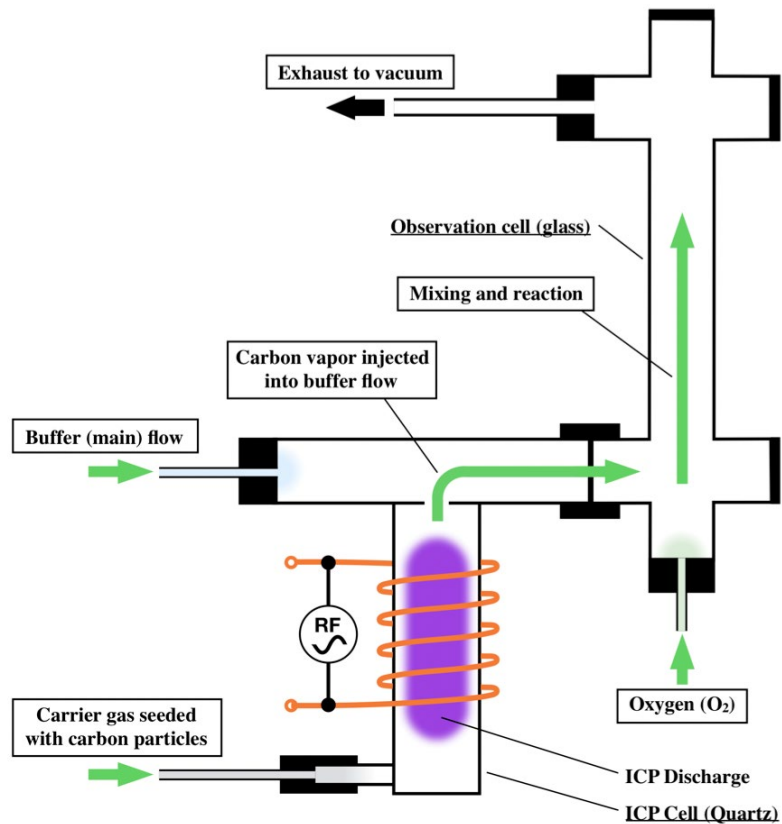
- $\text{N}_2(\text{A}^3\Sigma)$ generated in uniform nitrogen plasma in CRDS cavity by a ns pulse discharge burst
- Absolute, time-resolved $\text{N}_2(\text{A}, v=0-2)$ after the burst measured by CRDS
- $\text{N}_2(\text{A}^3\Sigma)$ decay time is very long (~ 1 ms), will survive from plenum to Mach 5 test section
- Measurements in Mach 5 CRDS cavity are underway

Related Work Originated from these Projects

(supported by Lockheed Martin SkunkWorks)

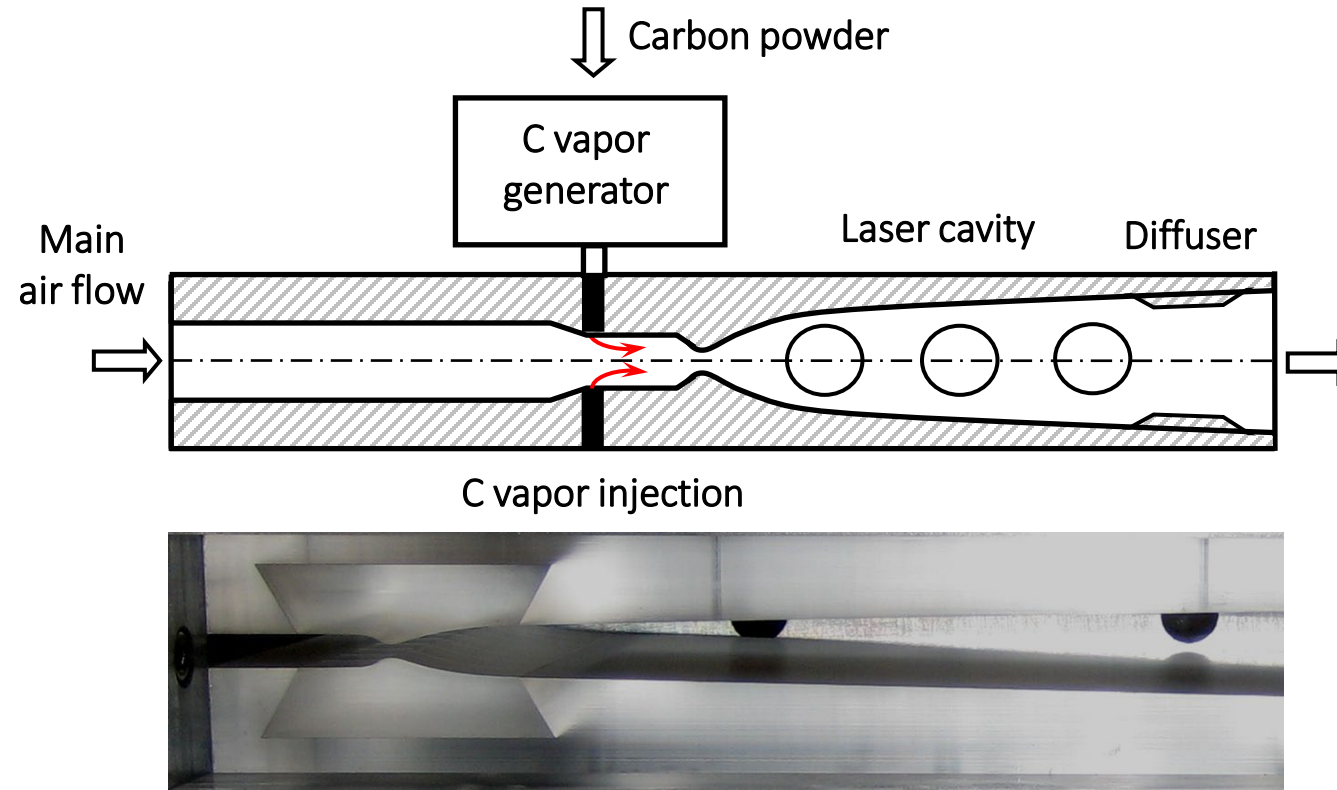
- Development of a novel supersonic flow chemical laser for electrical power generation on board of a hypersonic air vehicle
 - Ablation of carbon from a high-temperature surface in a hypersonic air flow
 - Reaction of C vapor with O₂ in the flow, generation of highly vibrationally excited CO
 - Creating population inversion, coupling out power in a CO laser resonator
 - CO lasers: scalable up to MW output power
 - Electrical power generation by photovoltaic conversion of laser power
- Approach to demonstrate feasibility in the lab
 - Carbon powder vaporized in a high temperature, inductively coupled plasma
 - Carbon vapor injected into airflow, reacts with O₂ in the flow: $C + O_2 \rightarrow CO(v) + O$
 - Demonstrate population inversion in CO product
 - Demonstrate laser action in a supersonic flow

Generation of C Vapor and Vibrationally Excited CO for a Novel Chemical Laser



- Micron size carbon particles seeded in Ar vaporized in the plasma
- Carbon vapor injected into airflow, reacts with O₂ in the flow: $\text{C} + \text{O}_2 \rightarrow \text{CO}(\text{v}) + \text{O}$
- CO product: total vibration population inversion, coupling out laser power is feasible

Goal: Demonstrate Lasing in a Supersonic Flow



- Generate carbon vapor in a high-temperature plasma, inject into airflow in plenum
- Carbon vapor reacts with O_2 in the flow: $C + O_2 \rightarrow CO(v) + O$
- Couple laser power in transverse supersonic flow resonator
- Supersonic flow laser infrastructure available in the lab

SUMMARY

- Development, operation, and instrumentation of a nonequilibrium flow wind tunnel, quantification of effect of vibrational relaxation on high-speed flow field
- Development of ps and ns CARS diagnostics to characterize vibrational nonequilibrium in the flow
- Development of pulse-burst laser / flow imaging system: portable diagnostics for high frame rate characterization of high-speed nonequilibrium flows
- Use of the pulse-burst laser for 500 kHz Mach 5 flow velocimetry at OSU, 500 kHz imaging of Mach 10 transitional boundary layer at NASA LaRC, 10 kHz imaging of flow in a model supersonic combustor in the 48" tunnel at CUBRC
- Development of close-coupled, state-specific vibrational-rotational-translational (V-R-T) rates for nonequilibrium NS and DSMC flow codes; models used extensively in the field
- Coordinated development of kinetic models, relevant experimental work, and model validation
- On-going work: state-specific measurements of nonequilibrium molecular dissociation, for validation of recently developed high-fidelity models
- On-going work: state-specific measurements of energy transfer from "dark" molecular states on UV radiation behind strong shocks
- On-going related work (support by Lockheed Martin SkunkWorks): development of a novel supersonic flow chemical laser for electrical power generation on board of a hypersonic air vehicle