Experimental characterization of energy transfer in nonequilibrium plasmas and high-speed flows using optical diagnostics

Igor V. Adamovich, <u>Walter R. Lempert</u>, J. William Rich,
Naibo Jiang, <u>Aaron Montello</u>, Munetake Nishihara, and Keisuke Takashima

Michael A. Chaszeyka Nonequilibrium Thermodynamics Laboratories
Department of Mechanical and Aerospace Engineering
The Ohio State University

The Michael A. Chaszeyka Nonequilibrium Thermodynamics Laboratory

The NETL Group

- 3 faculty (appointments in Mechanical and Aerospace Engineering, Chemical Physics, and Chemistry)
- ~ 15 graduate students, post-docs, visiting scholars. Backgrounds in engineering, physical chemistry, plasma physics

Objectives / Outline

- **Obtain experimental data on molecular energy transfer mechanisms / rates to enable predictive modeling of high-speed nonequilibrium flow fields**
- **Develop instrumentation to measure temperature, vibrational populations, species concentrations: high frame rate NO PLIF, high frame rate NO_2 MTV, psec CARS, TALIF, Thomson scattering**
- **Demonstrate use of such instrumentation to obtain data in short duration hypersonic flow facilities (shock tunnels)**
- **Develop methods of actively influencing flow field energy storage, energy transfer, and aerodynamic control**
- **Test bed I: a small scale Mach 5 nonequilibrium flow wind tunnel, with vibrational energy of air species loaded by an electric discharge in plenum, controlled by adding relaxer species downstream**
- **Test bed II: nsec pulse, point-to-point, filament discharge loading vibrational energy in quiescent N_2 or air**
- **Kinetic modeling: do we really understand energy transfer mechanisms involved?**

Test Bed I: Mach 5 Nonequilibrium Flow Wind Tunnel
Operating Conditions

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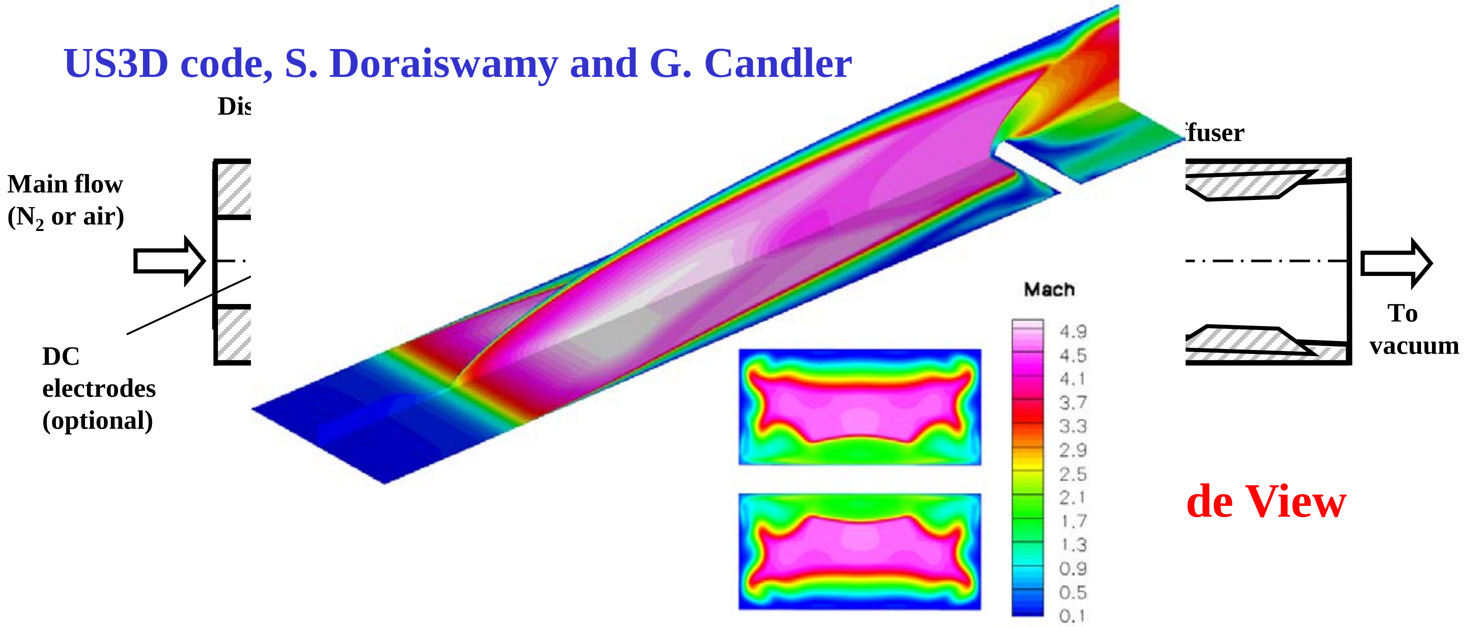
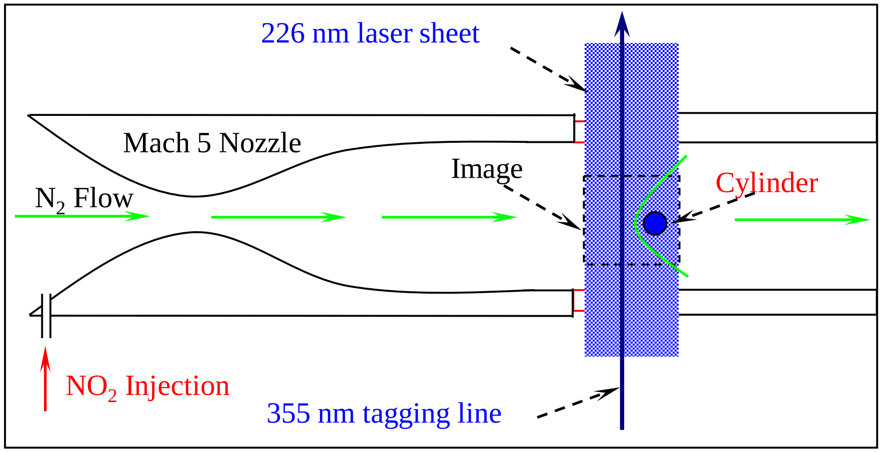
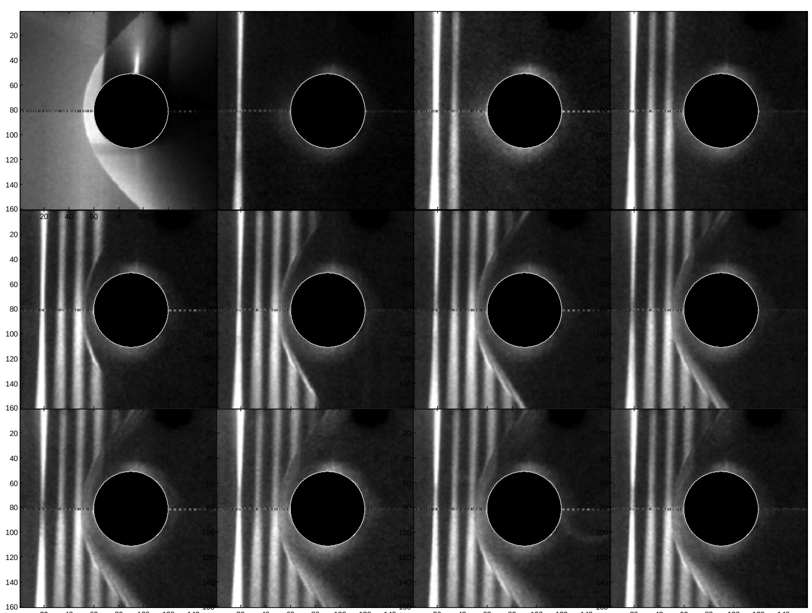


- Mach 5, steady state run time 5-10 seconds, P_0=0.5-1.0 atm (M~0.25 flow in plenum)
- Supersonic test section: 4 cm x 4 cm cross section, flow over a 5 mm cylinder model, 4 optical access windows for optical diagnostics
- Sustaining nonequilibrium flows: vibrational energy loading by nanosecond pulser / DC sustainer discharge in plenum
- Discharge power up to ~ 3 kW, power density ~ 300 W/cm^3
- Controlling vibrational disequilibrium: injection of V-V / V-T relaxers (O_2, NO, CO_2, H_2) downstream of discharge / upstream of the throat

Nishihara et al., AIAA J., 2012

Previous work: 500 kHz NO PLIF and Molecular Tagging Velocimetry in Mach 5 wind tunnel (equilibrium flow)

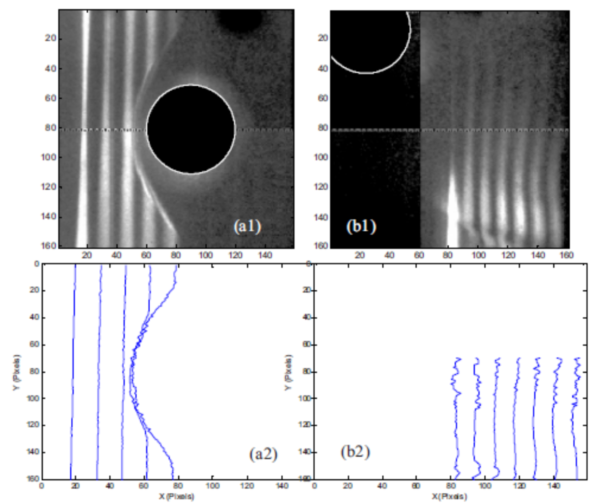




NO_2 MTV Method

Tag: $NO_2 + h\nu \rightarrow NO + O$

Interrogate: **NO PLIF Imaging**



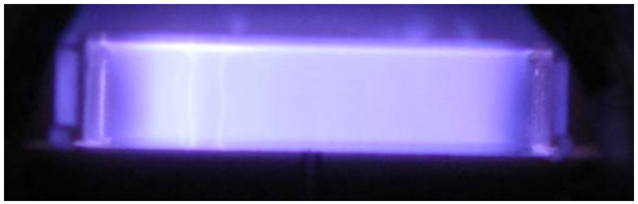
Free Stream Results

$$\overline{V} = 719\, m/s, \qquad \sigma_V = 10\, m/s$$

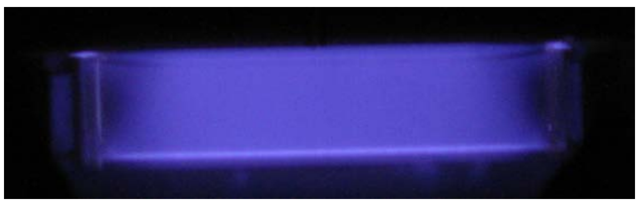
Jiang et al, Appl. Phys. Lett., 2011

Nsec Pulse Discharge:
Large-Volume Diffuse Ionization

Flow into the screen

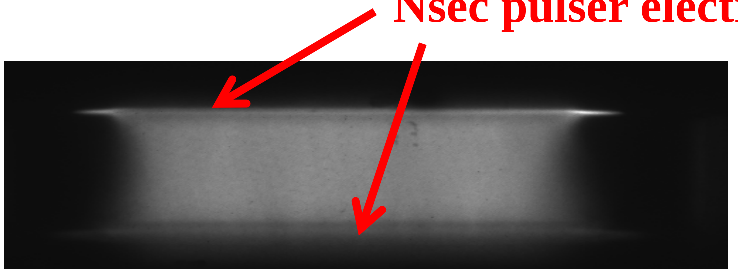


Nitrogen, P_0 = 300 Torr, ν = 100 kHz, no DC electrodes

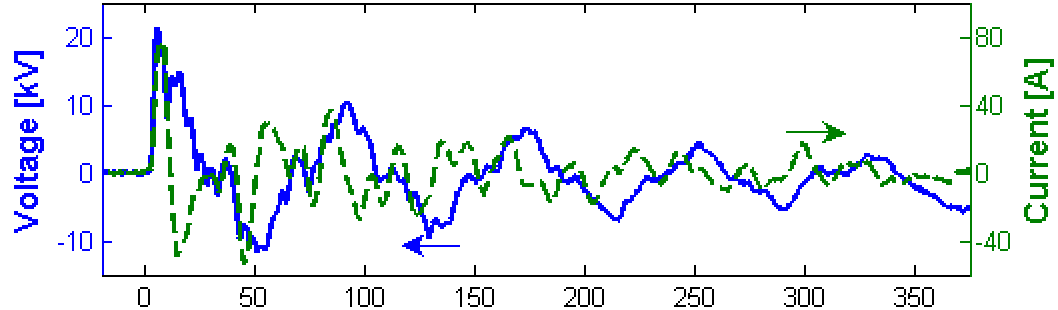


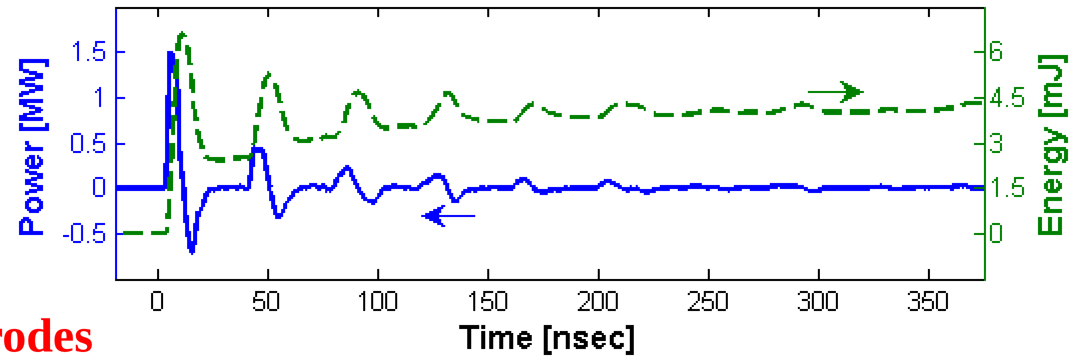
Nitrogen, P_0 = 650 Torr, ν = 100 kHz, no DC electrodes

Nsec pulser electrodes



Nitrogen, P_0 = 350 Torr, ν = 100 kHz, 1000th pulse, 5-µsec camera gate





Nitrogen, P_0 = 300 Torr,
ν=100 kHz, DC electrodes not powered
0.1 sec after beginning of burst (i.e. pulse # 10,000)

Pulse energy 4.5 mJ/pulse
Average nsec pulser power 450 W

Nishihara et al., AIAA J., 2012

Energy Addition: DC Sustainer Discharge

(overlapped with nsec pulse discharge in nozzle plenum)

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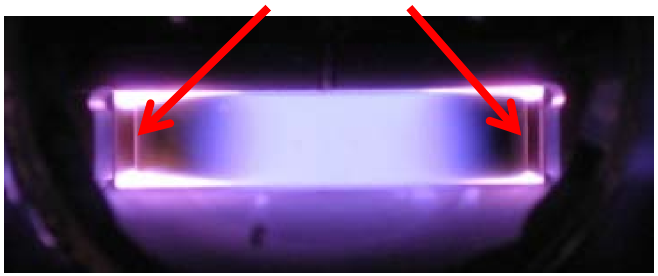
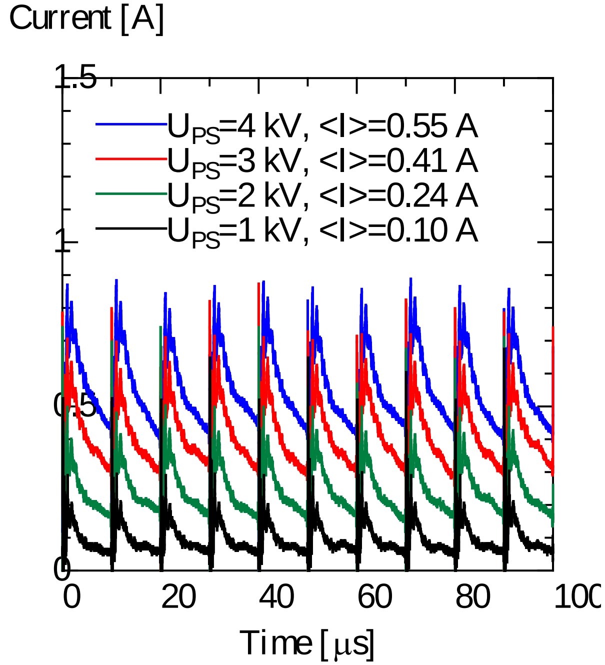
DC sustainer electrodes

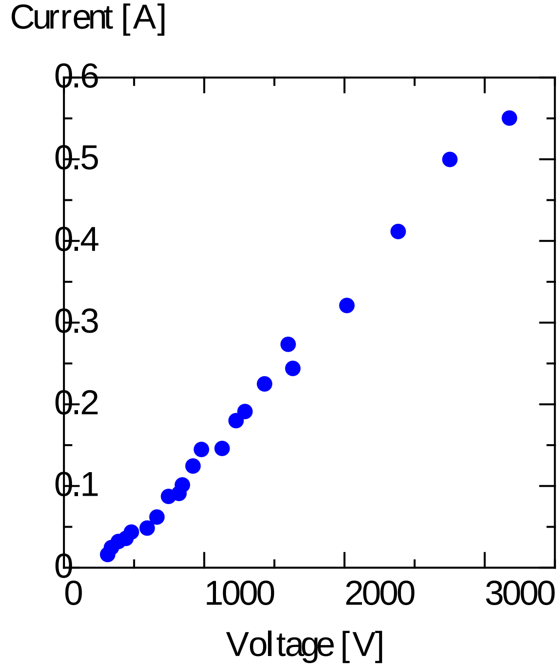


U_{PS} = 2 kV , U = U_{PS} - IR = 1.5 kV

Nitrogen, P_0 = 350 Torr, ν = 100 kHz:

- Plasma does not fully decay between nsec pulses
- Linear current rise with voltage: non-self-sustained DC discharge
- DC sustainer discharge stable up to 3 kW (U_{PS}=3.5 kV)
- T_0=400 K (N_2 emission), T_{v0}=1800 K (estimated)





Nishihara et al., AIAA J., 2012

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Life before CARS: 2-line NO PLIF thermometry in Mach 5 flow over a cylinder model

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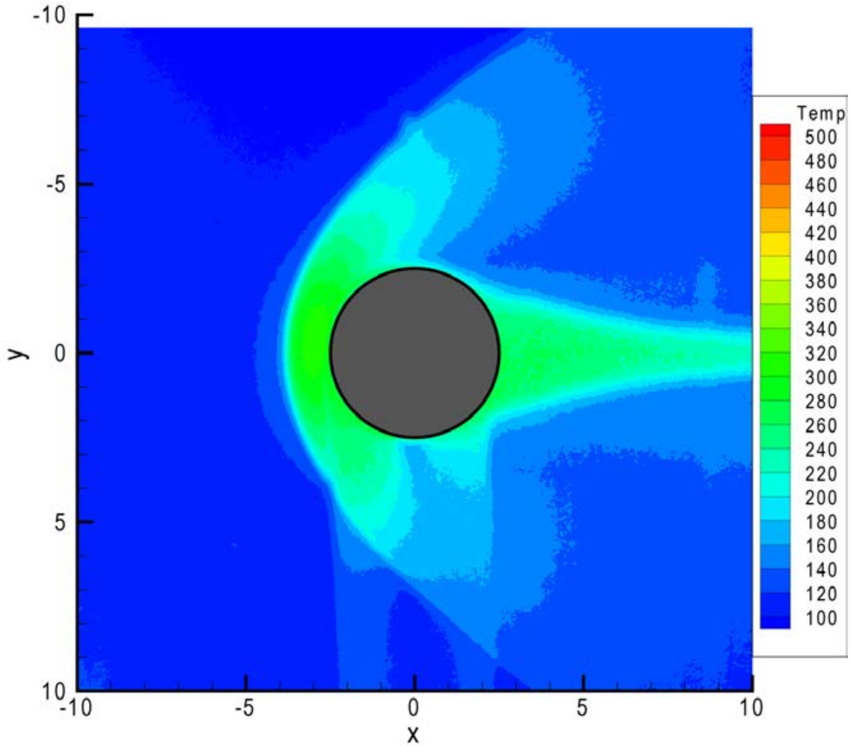
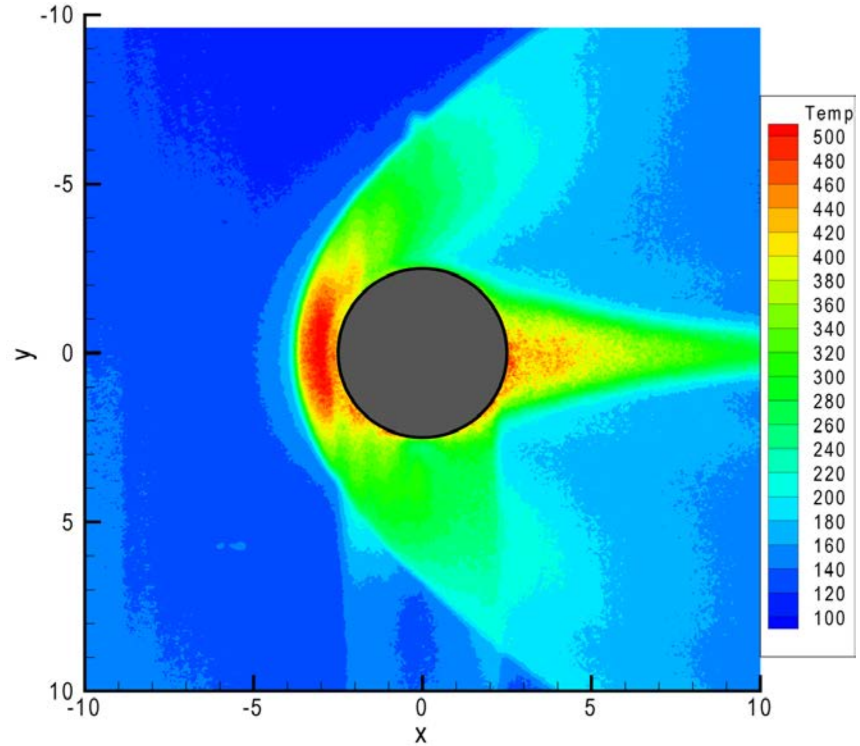


Rotational temperature distribution without discharge in plenum. Nitrogen, P_0=370 torr + 0.3 torr NO.



Rotational temperature distribution with pulsed/DC discharge in plenum (ν=100 kHz, U_{PS}=4.5 kV).

What is $T_v(N_2)$?

Nishihara et al., AIAA J., 2012

Coherent Anti-Stokes Raman Scattering (CARS) Spectroscopy – Basic Principles

CARS four wave mixing process can be thought of as two 2-photon processes:

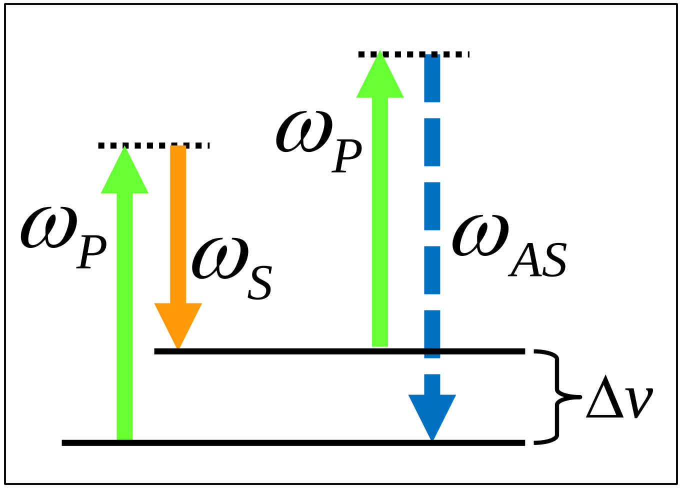
(1) Pump/Stokes photons induces a coherent (i.e. phased) oscillating polarization

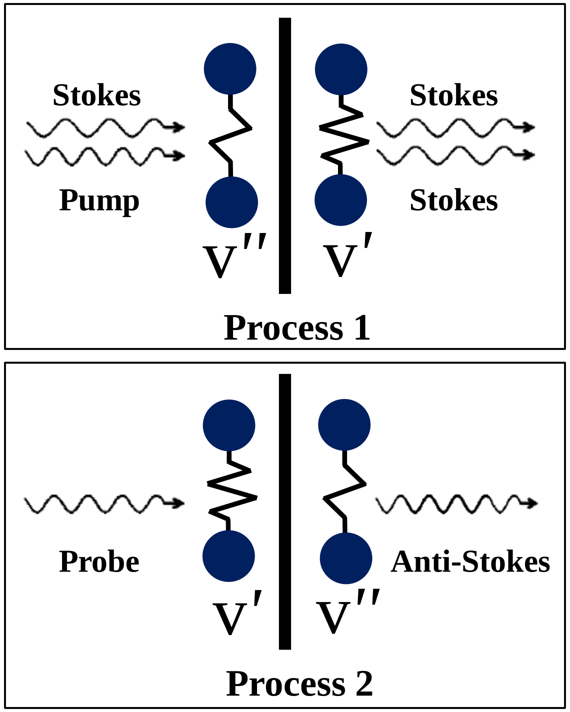
For N_2 vibrational CARS, a 532 nm pump requires a 607 nm Stokes beam

(2) Probe photon induces anti-Stokes Raman scattering, coherent in the phase matching direction

$$I_{CARS} \propto I_{pump} \cdot I_{Stokes} \cdot I_{probe} \cdot n^2$$

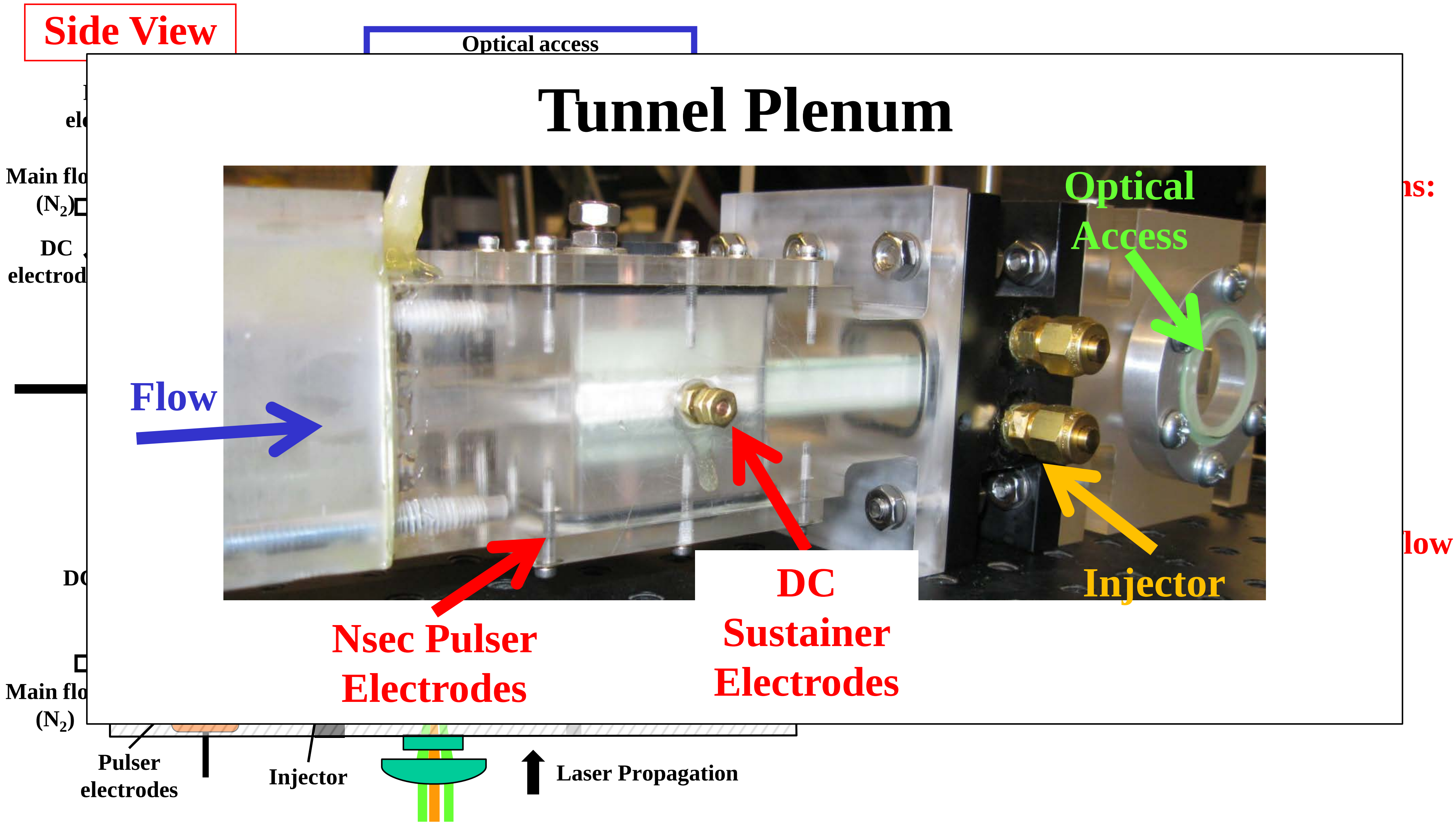
CARS Energy Level Diagram





Psec CARS: higher signal –to-noise (single-shot spectra possible), sub-nsec time resolution, no optical window damage

CARS Diagnostics in Mach 5 Nonequilibrium Wind Tunnel



Collinear CARS Schematic:
Measuring $N_2(X,v)$ populations in wind tunnel plenum

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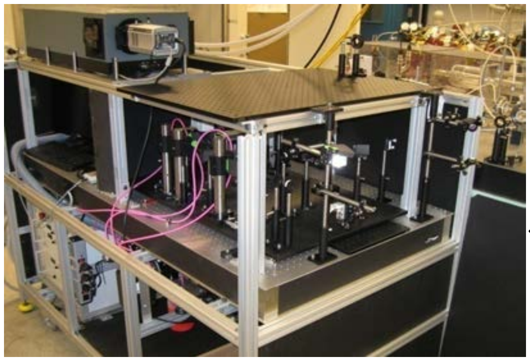

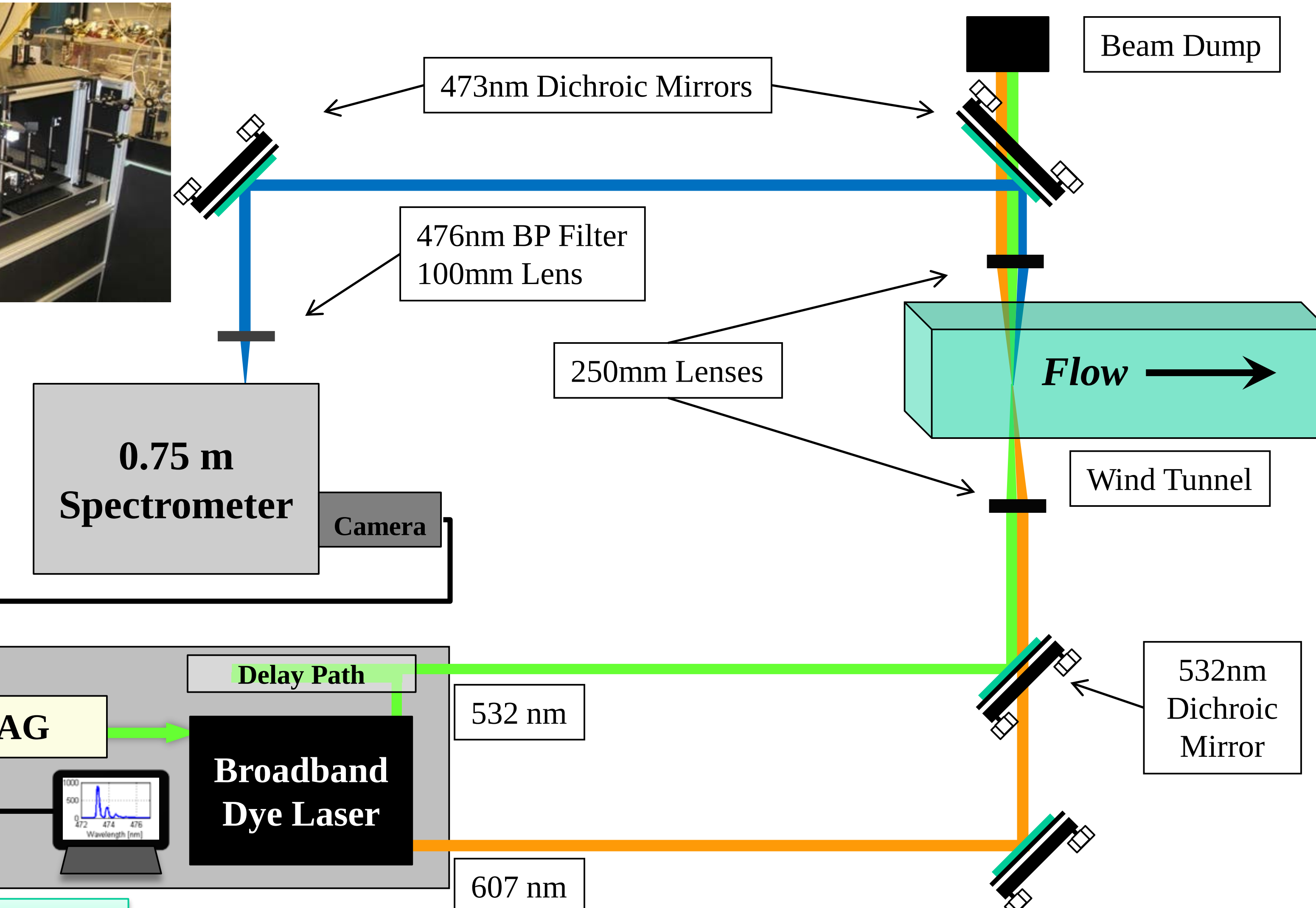

Montello et al, AIAA J., 2012

Broadband Dye Laser (“Stokes”) Spectrum:
Simultaneous access to multiple vibrational states

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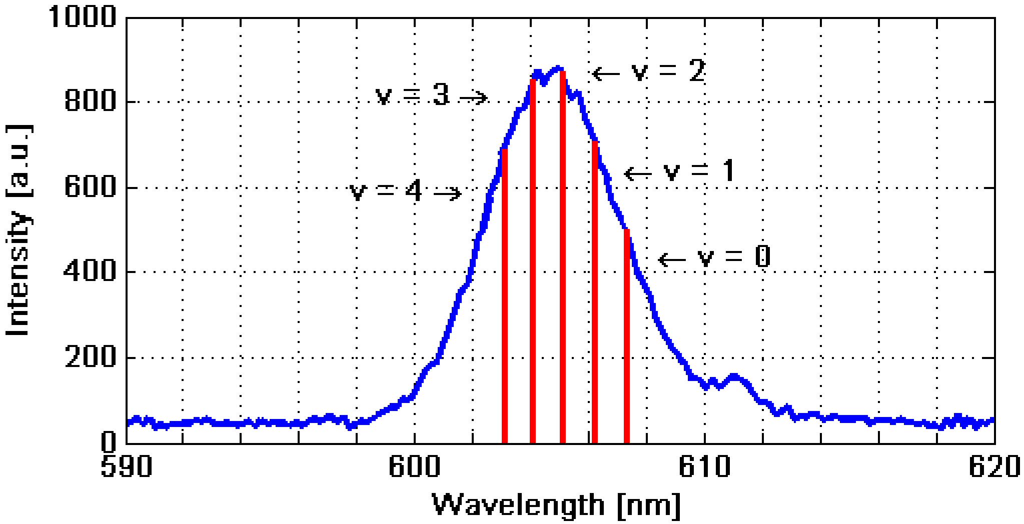

$$\frac{E_v}{h} = \omega_e(v+\tfrac{1}{2}) - \omega_e x_e (v+\tfrac{1}{2})^2$$

$$\frac{\Delta E_v}{h} = \omega_e - 2\omega_e x_e (v+\tfrac{1}{2})$$

Vibrational energy of a diatomic molecule (ignoring rotation)

Broadband dye laser spectral profile, with necessary Stokes frequencies superimposed: enough bandwidth to probe several levels simultaneously

Transition	Raman Shift [cm^{-1}]	Stokes wavelength [nm]	Anti-Stokes wavelength [nm]
0 – 1	2330.7	607.3	473.3
1 – 2	2301.8	606.2	474.0
2 – 3	2272.9	605.2	474.6
3 – 4	2244.0	604.1	475.3
4 – 5	2215.1	603.1	475.9

For N_2:

$$\omega_e = 2359.61\,\text{cm}^{-1}$$
$$\omega_e x_e = 14.456\,\text{cm}^{-1}$$

Single-Shot N_2 CARS spectra in 300 Torr N_2: Pulser Alone vs. Pulser/Sustainer

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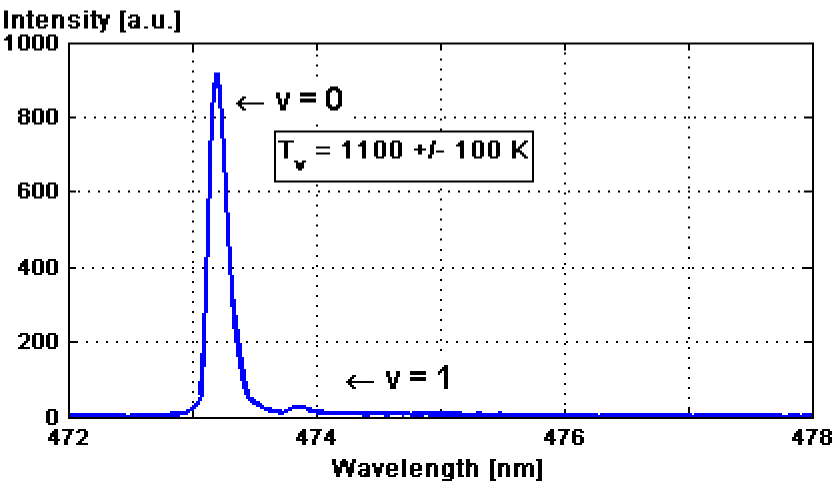
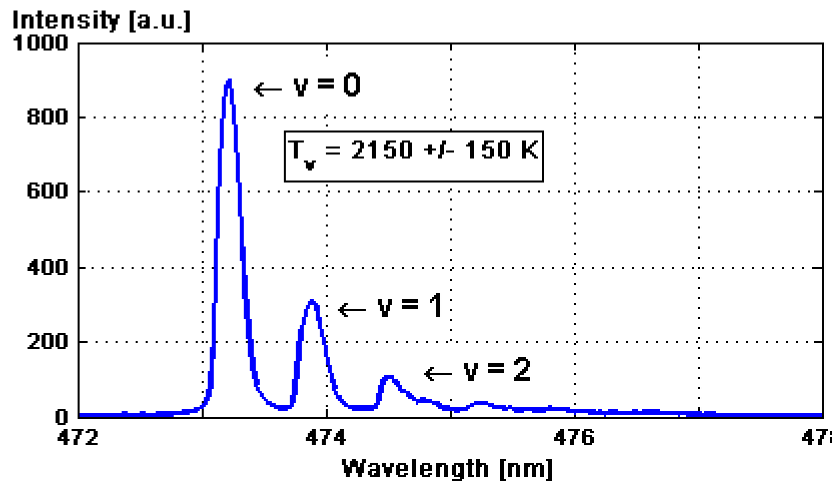
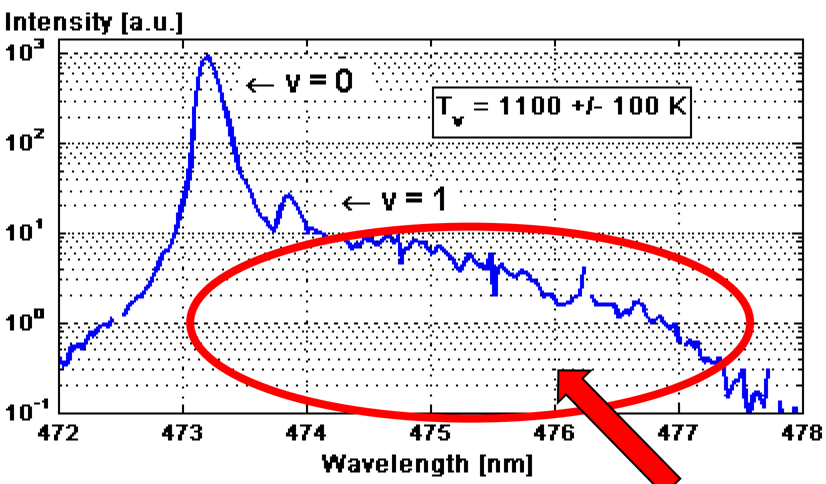
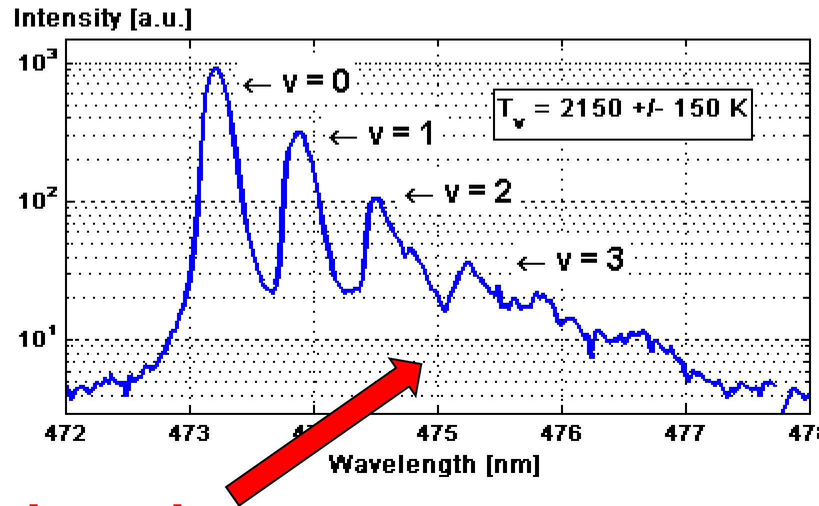


Nanosecond pulser alone



Pulser-Sustainer discharge, 4.5 kV DC





Non-Resonant Background

Montello et al, AIAA J., 2012

Time-resolved $N_2(X)$ vibrational temperature in a nsec pulse / DC sustainer discharge

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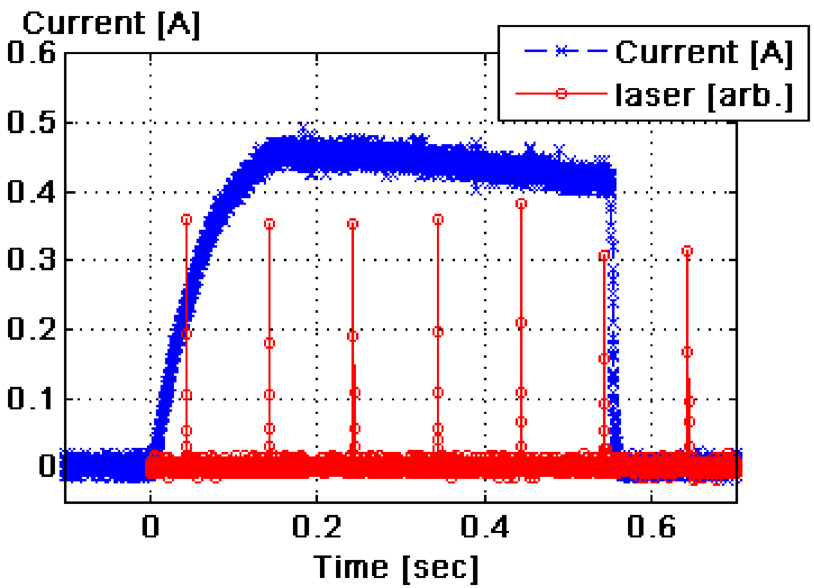

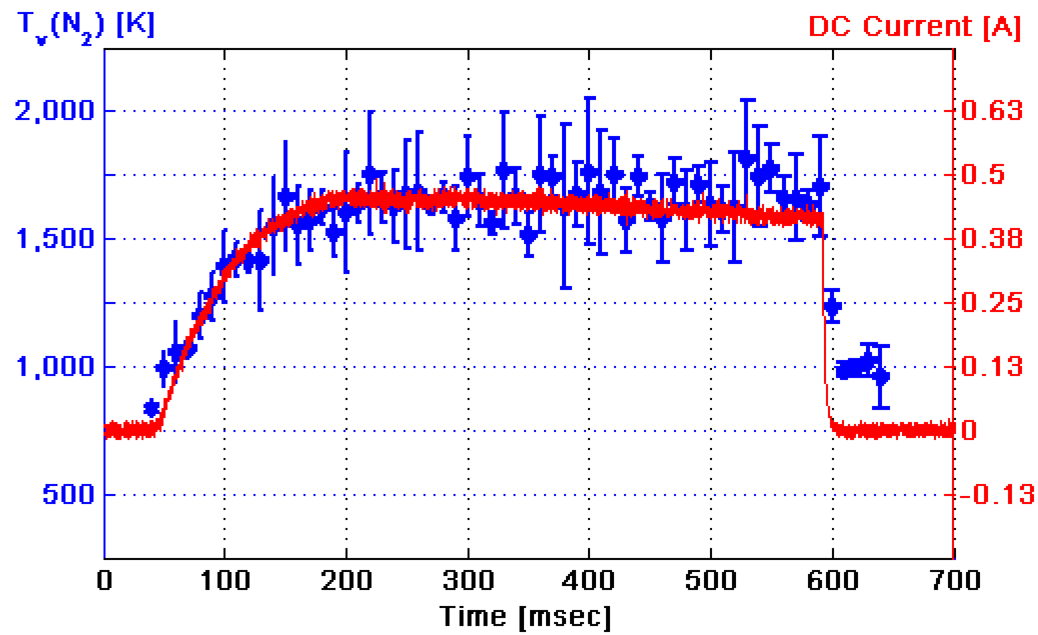

- Varied laser delay timing to map out temporal evolution of $T_v(N_2)$
- $T_v(N_2)$ was observed to directly follow DC sustainer current profile

- Low T_v system threshold ~ 800-1000 K observed (corresponds to nsec pulser excitation w/o DC sustainer)
- Threshold can be improved by reducing non-resonant background

Montello et al, AIAA J., 2012

Schematic of USED-CARS Experimental Arrangement

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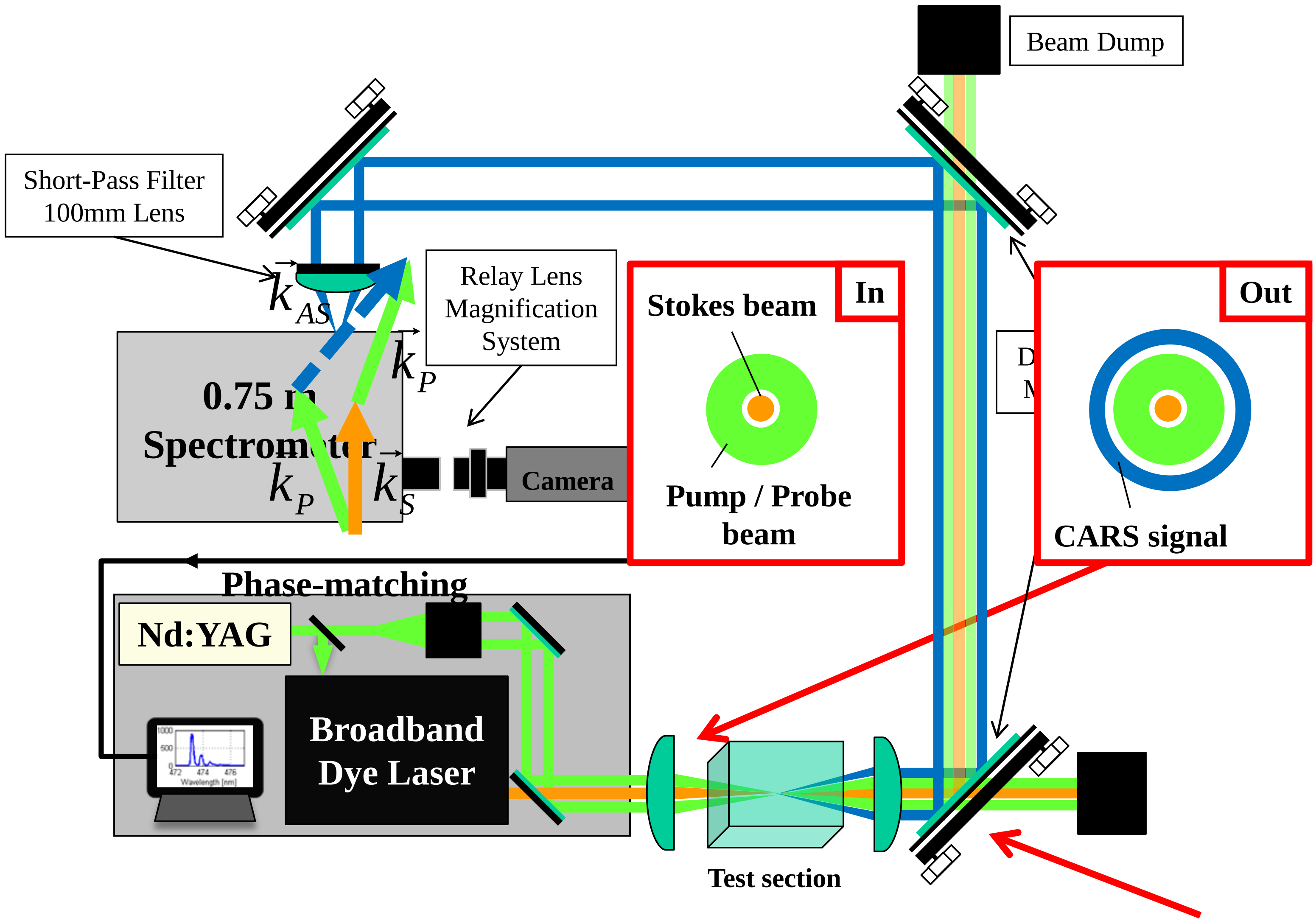

Rotational / Vibrational CARS Spectra:
Pulser-Sustainer Discharge – with / without relaxant injection

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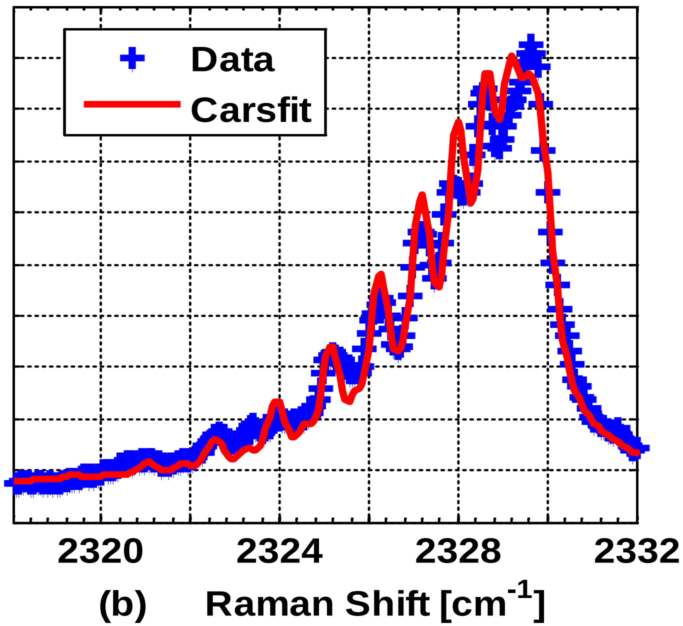

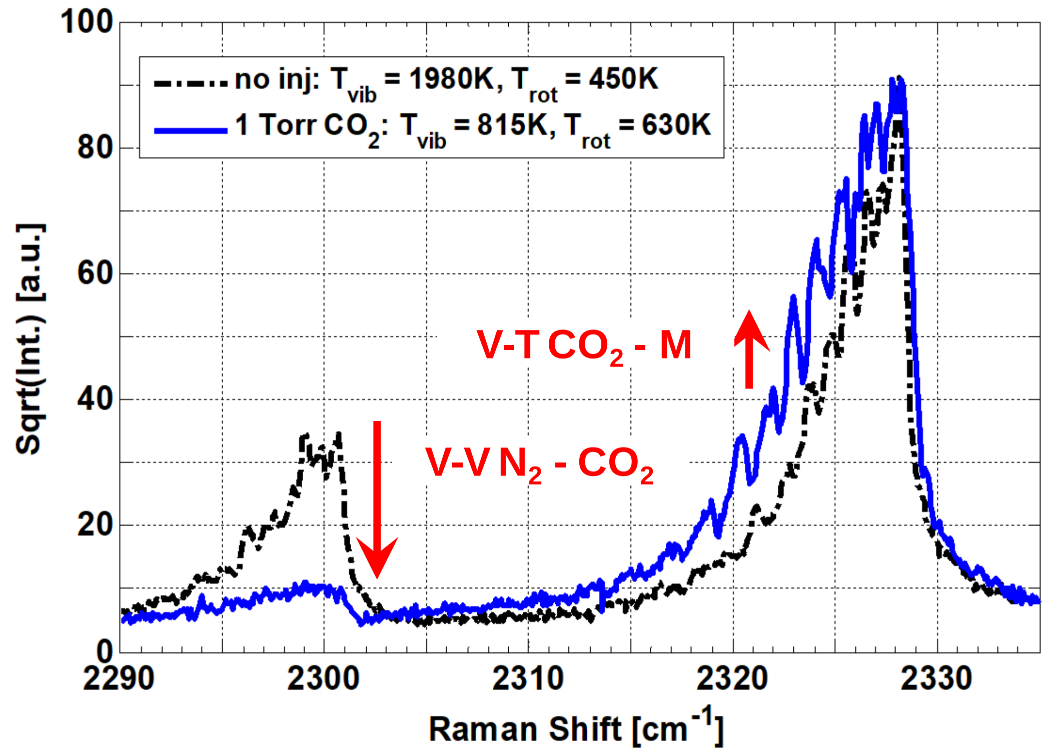

20-shot average spectrum
No discharge, T=300 K, P_0=300 Torr
With Sandia CARSFIT synthetic spectrum
T_{fit} = 322±10 K.

20-shot average spectra, P_0=300 Torr:
Pure N_2 and N_2 with 1 Torr CO_2 partial pressure

Pulser-Sustainer Discharge:
Effects of V-V / V-T Relaxer Injection

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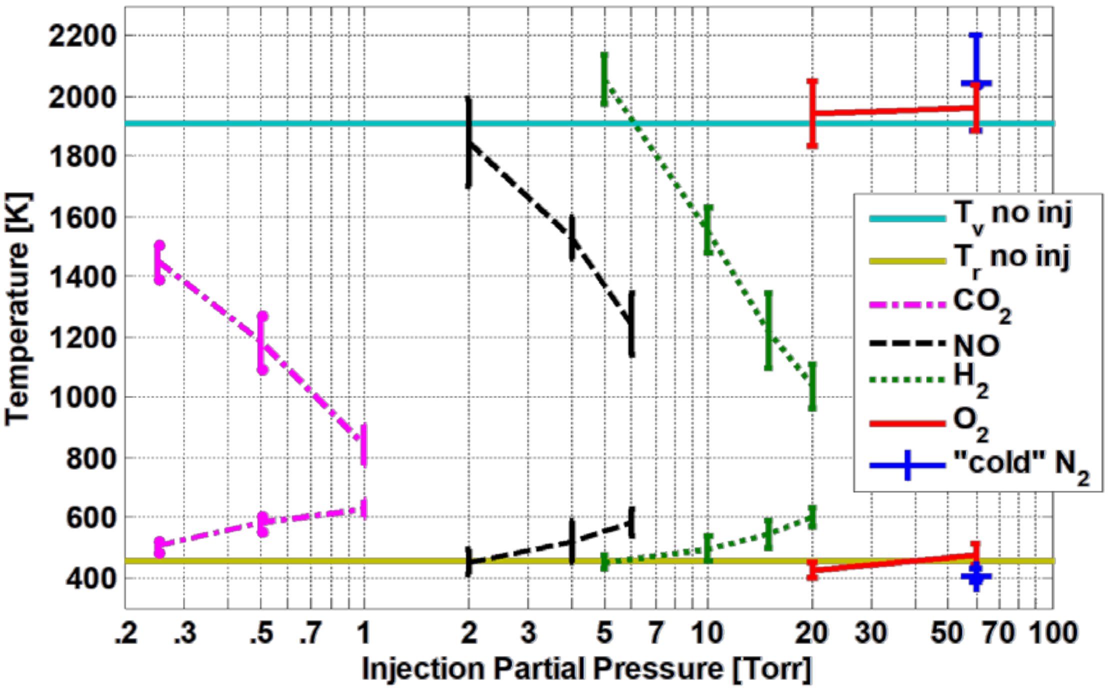


Characteristic relaxation times (from kinetic rates – $\tau_{VV} \sim 1/k_{VV}n_{add}$):

1 Torr CO_2 (V-V) ~ 70 µsec

5 Torr NO (V-V) ~ 2 msec

10 Torr H_2 (V-T) ~ 6 msec

60 Torr O_2 (V-V) ~ 17 msec

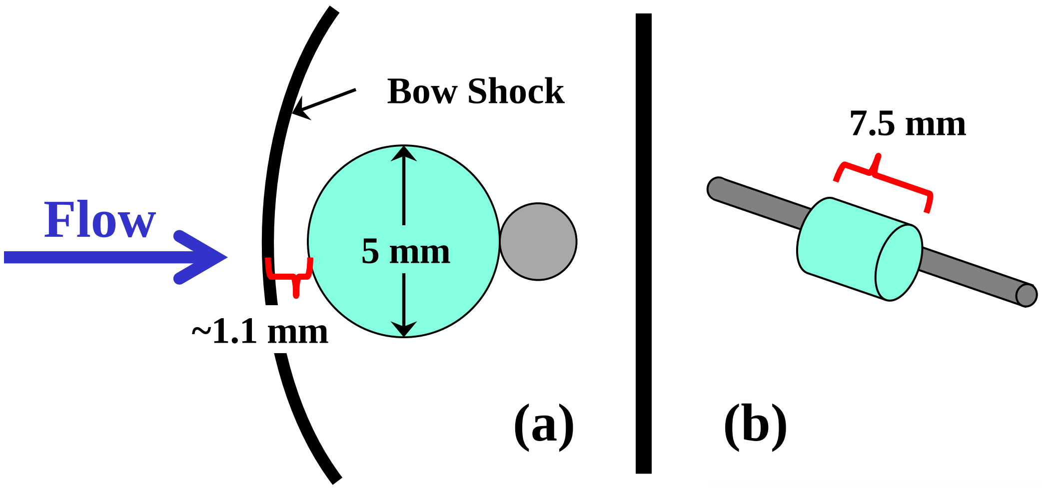
Flow residence time ~ 2 msec

300 Torr total mixture pressure
Injection of CO_2, NO, H_2, O_2 and N_2

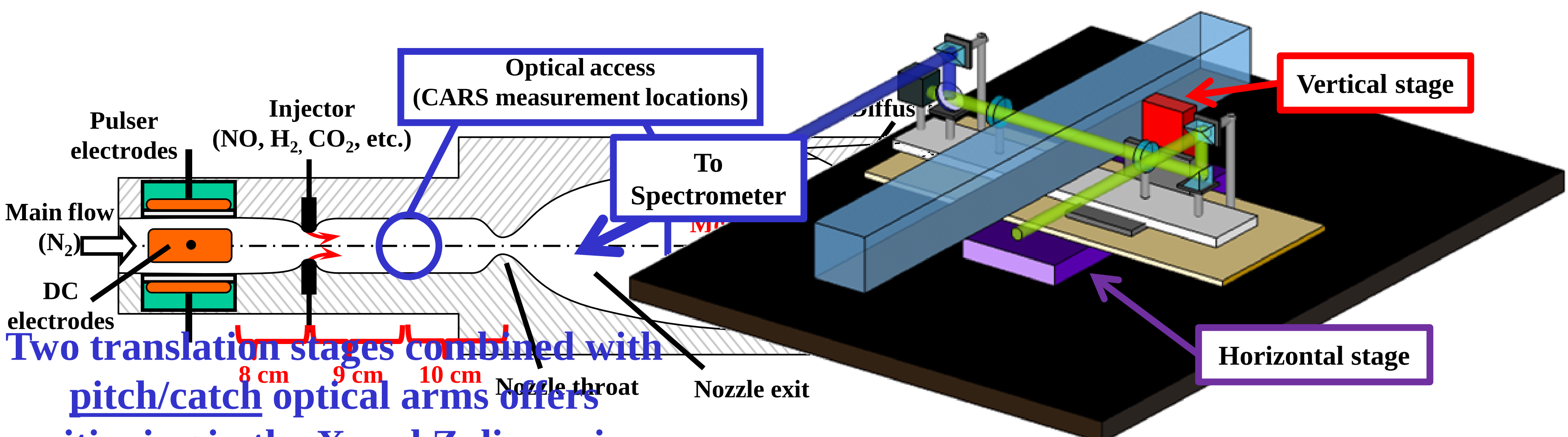
No effect of O_2 addition on T_v (N_2) due to slow N_2-O_2 V-V energy transfer

E_{vib} (N_2) + $E_{trans/rot}$ (all species) ~ const (no vib. energy storage in relaxer species)

Schematic of 2-D CARS measurements in Mach 5 flow (XZ-resolution)



5 mm diameter x 7.5 mm long quartz cylinder model attached to 2 mm stainless steel support rod, creates bow shock in the Mach 5 flow



Two translation stages combined with <u>pitch/catch</u> optical arms offers positioning in the X and Z dimensions

CARS Spectra in Mach 5 Flow

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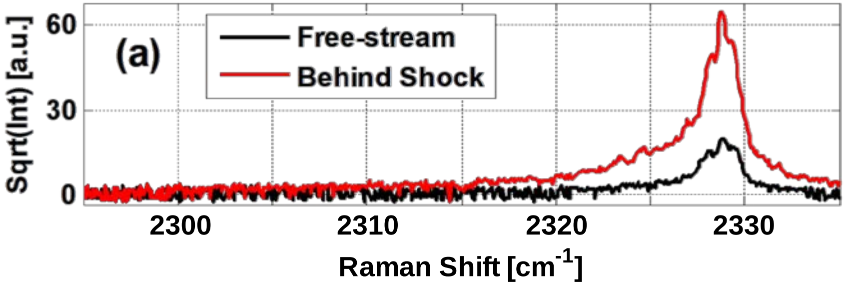

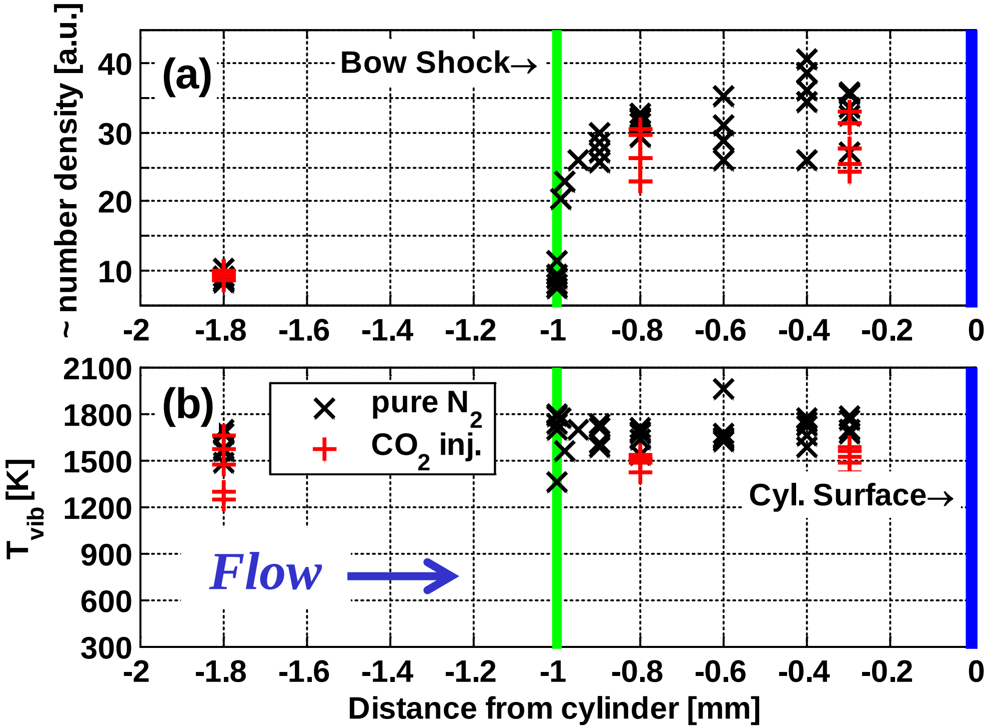

(a) 8-shot averages - freestream (P=1.2 Torr, T ~ 50 K) and behind the shock (P~ 30 Torr, T~ 300 K) -- Plasma OFF

(b) 10-shot average, supersonic freestream -- Pulser-sustainer discharge

(c) 10-shot average, behind the bow shock -- Pulser-sustainer discharge

Change in number density indicates shock stand-off distance of 1.0 mm

Data collected within 300 µm of model surface

No detectable N_2 relaxation behind the shock

Test Bed II: Diffuse Filament Nsec Pulse Discharge between two bare metal spherical electrodes

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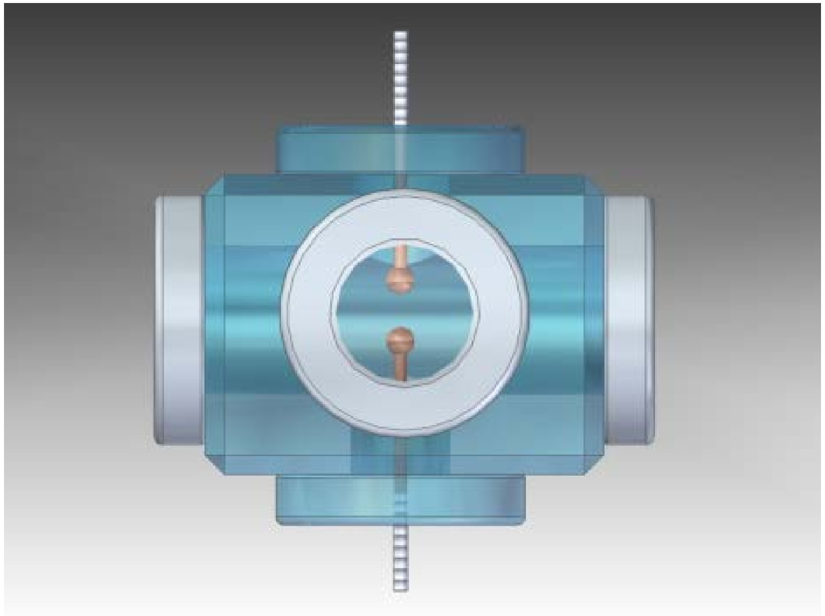
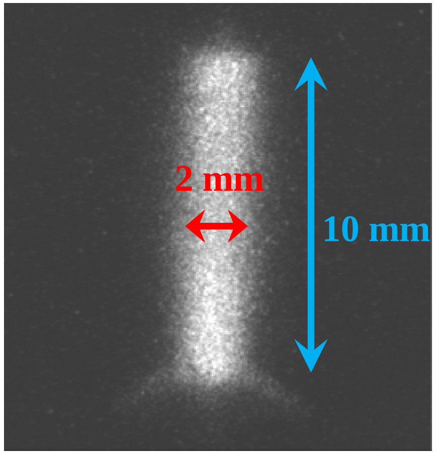
Use of small (a few mm diameter), bare spherical electrodes increases power loading (~0.3 eV/molecule/pulse at P=100 Torr, coupled pulse energy ~15 mJ)

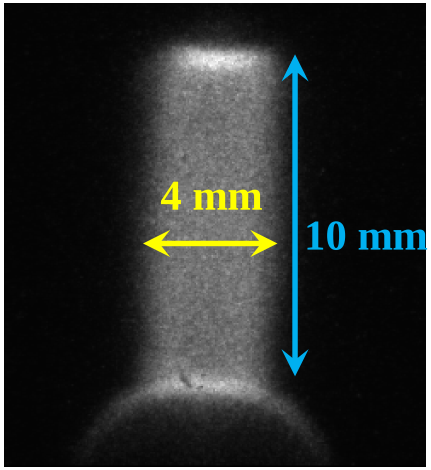
AND creates plasma large enough to be easily probed by CARS



N_2, 100 Torr
"low current" regime



Air, 100 Torr
"high current" regime



BOX-CARS, broadband dye mixture:
better spatial resolution, access to more vibrational levels

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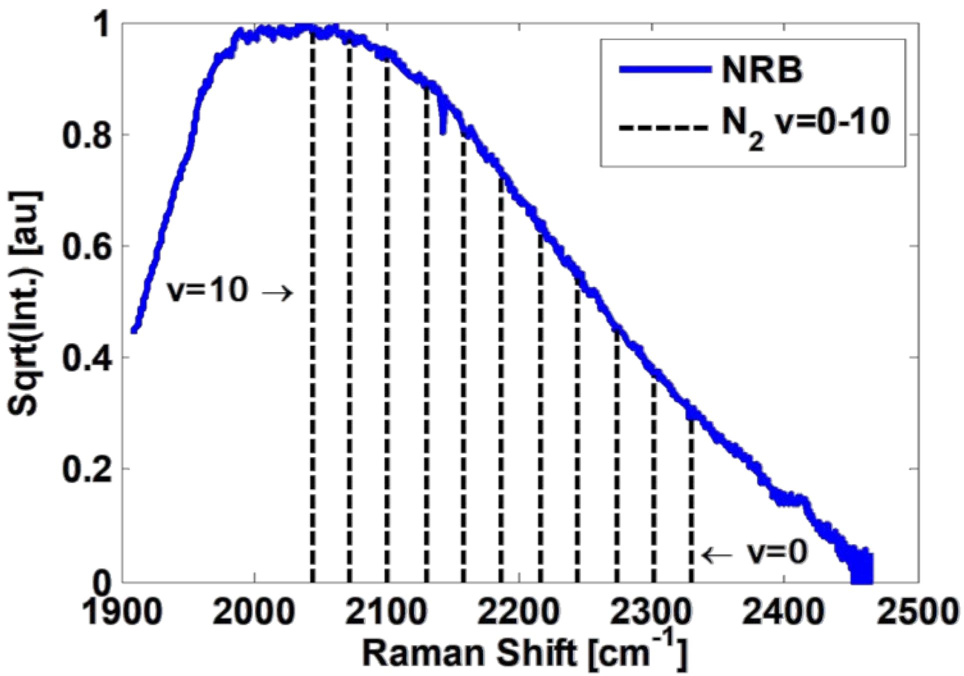
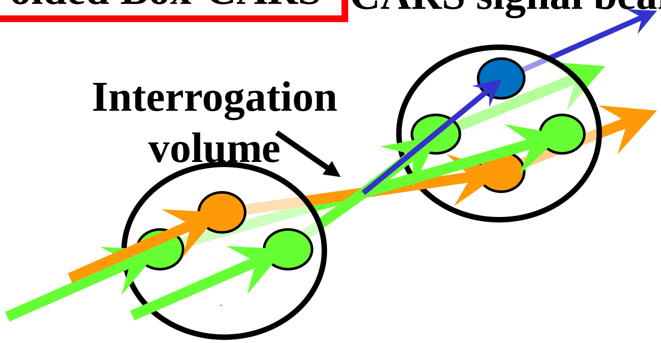
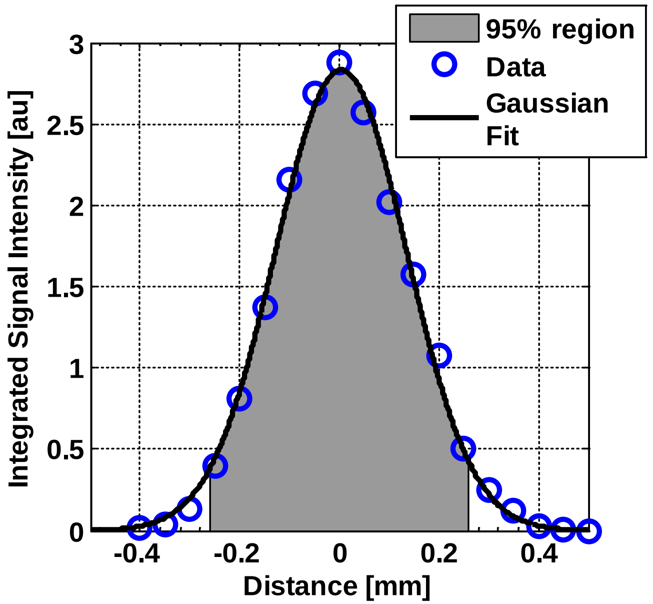
Very Broadband Pyrromethene Dye Mixture (*S. Tedder, et al, 2011)



Folded Box-CARS



95% of signal generated over ~0.5 mm



Characterization of discharge filament size:
N_2 in "low current" regime (U_{DC}=430 V)

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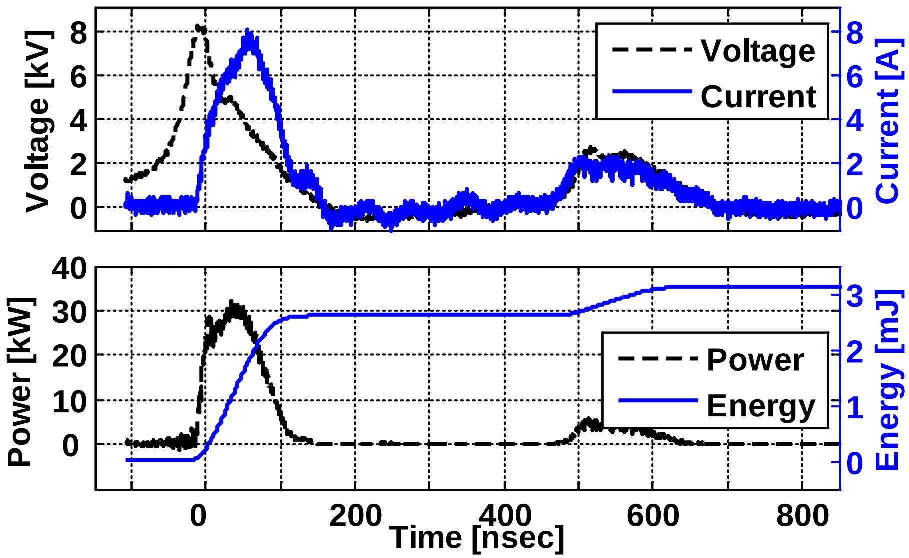
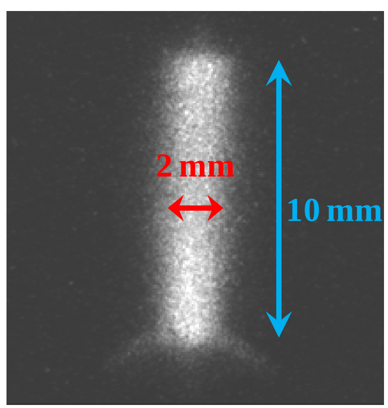


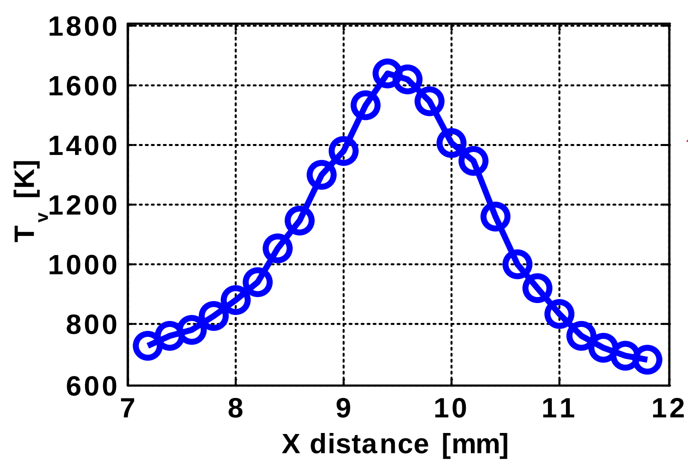
Voltage, current, power, and energy for "low lurrent" pulse discharge in N_2 (430 V DC)

Spatial scan of $T_v(N_2)$ across the discharge filament, consistent with ICCD measurements, FWHM ~2 mm

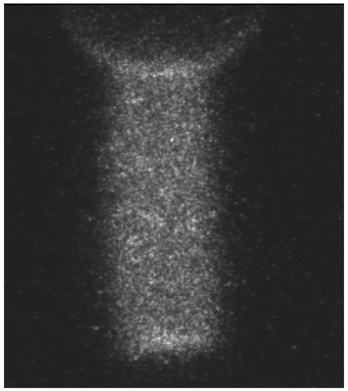
ICCD image of discharge filament

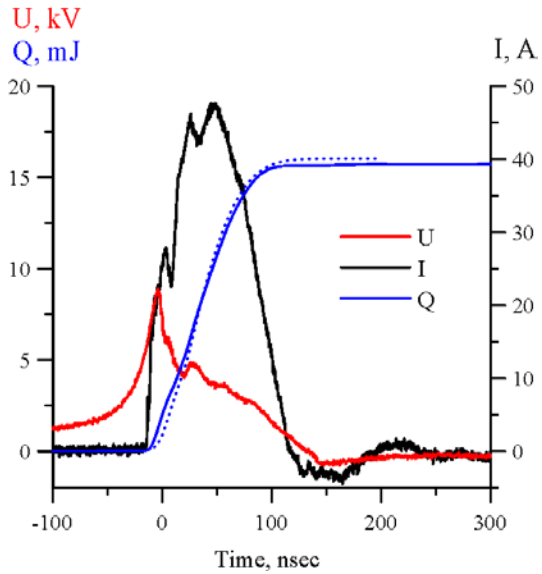
FWHM ~2mm

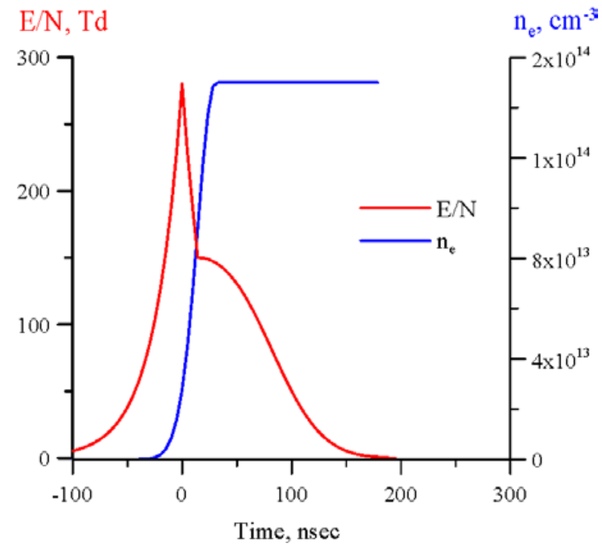


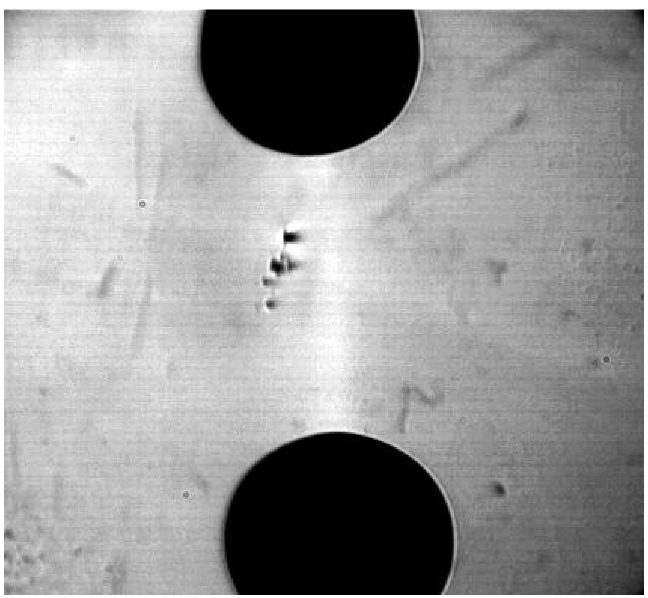


Air: images and waveforms in "high current" regime (U_{DC}=486 V)









Compression waves formed by "rapid" heating, on sub-acoustic time scale

From known initial temperature & pressure, voltage, current, and filament diameter →

Reduced electric field (E/N) and electron density for kinetic modeling

Typical CARS Spectra, 100 Torr N_2

(Normalized to v=0, corrected for dye laser spectral profile)

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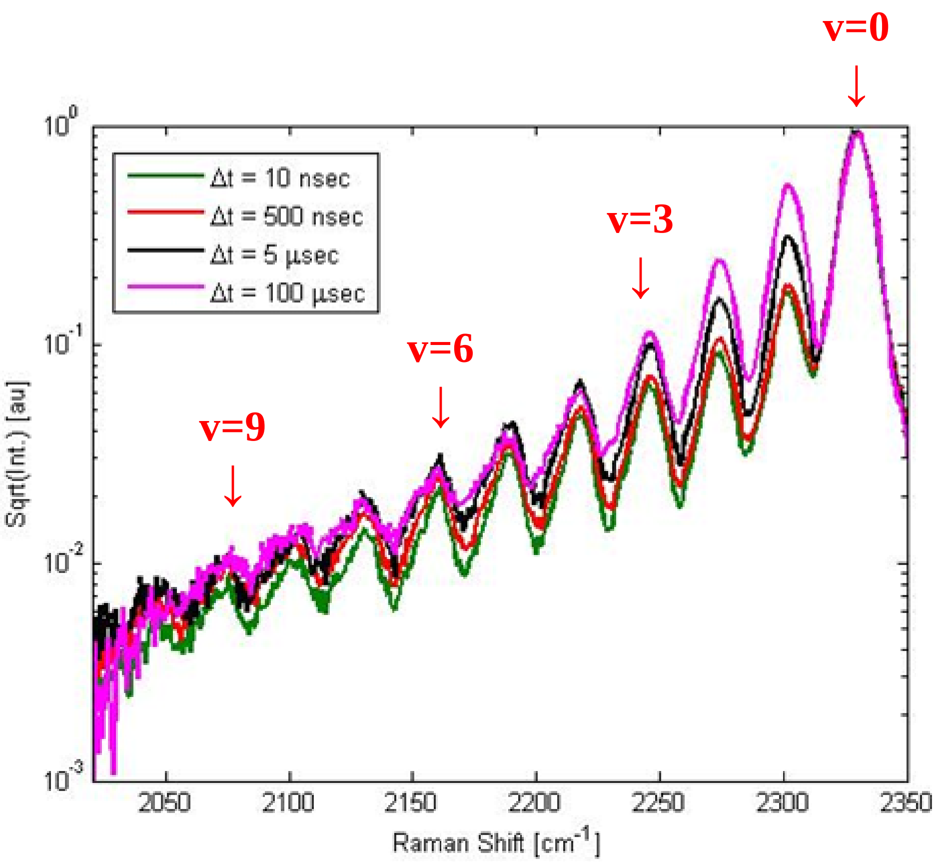
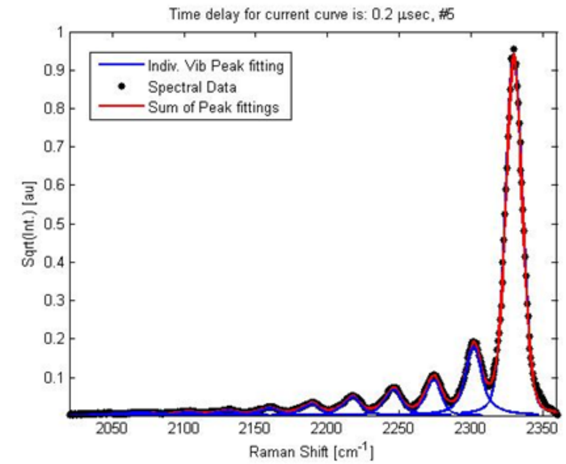
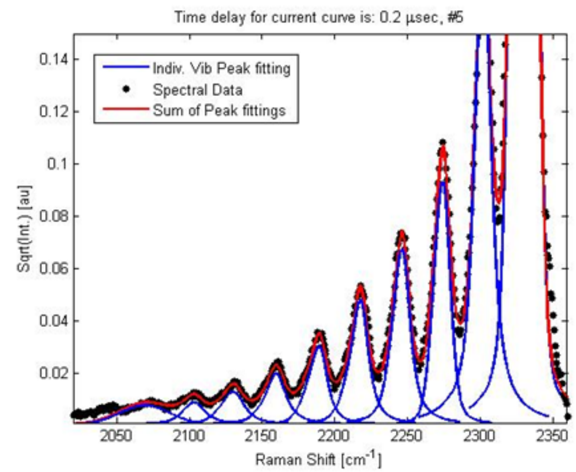
100 laser “shot” averaged spectra vs. time after rising edge of current pulse



Vibrational level populations inference: least squares fitting to Voigt line shape





Kinetic modeling: coupled master equation / Boltzmann equation model of nonequilibrium air plasma

Nonequilibrium Thermodynamics Laboratories

$$\frac{dn_v(t)}{dt} = (El.imp)_v + (VT)_v + (VV)_v + (VE)_v + (V-Chem)_v + Diffusion$$

RHS terms represent vibrational quantum state change by the following processes:

El. Imp.: inelastic electron impact processes by free electrons

VT, VV: vibration-to-translation/rotation relaxation, vibration-to-vibration energy exchange

VE: electronic-vibration energy transfer during collisional quenching

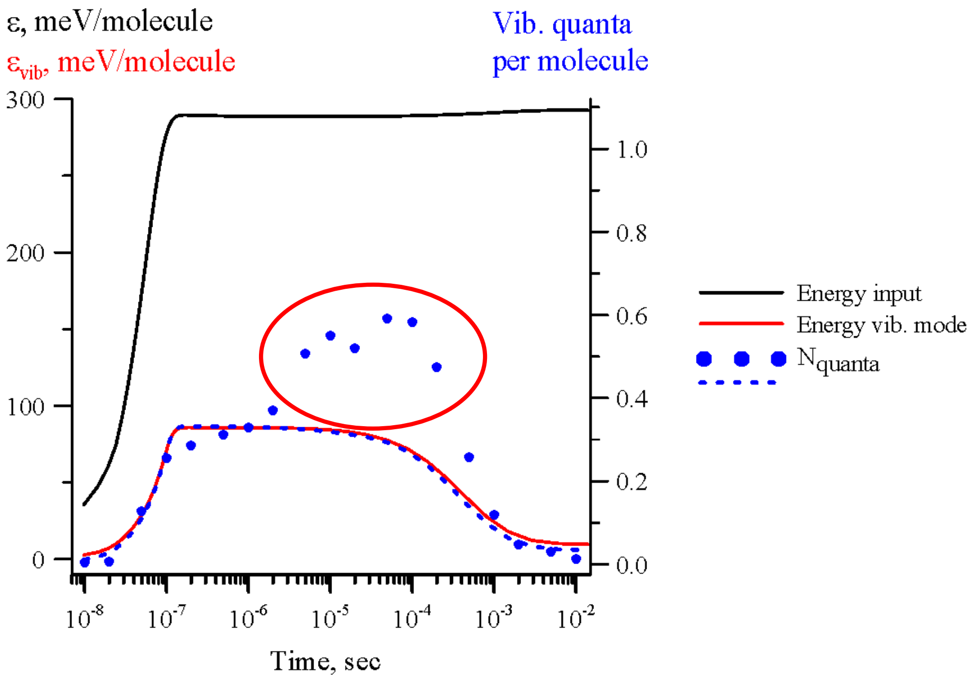
V-Chem: vibrational – chemistry coupling for vibrationally enhanced reactions such as

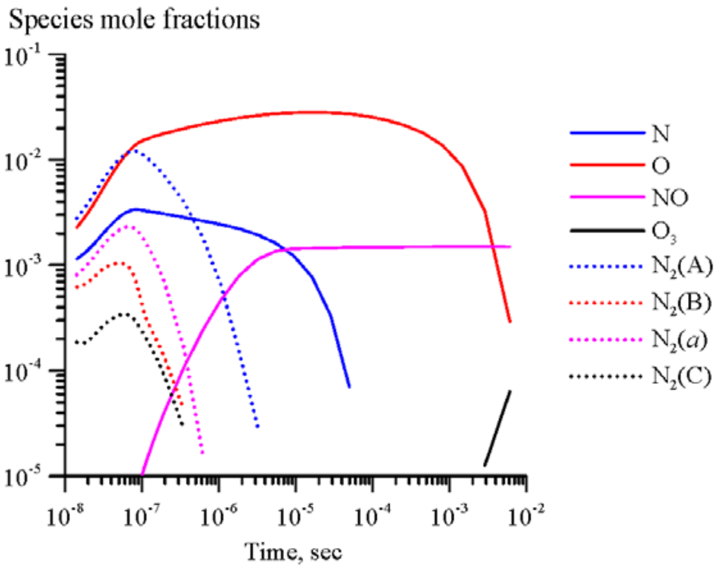
$N_2(v) + O \rightarrow NO + N$, $O_2(v) + N \rightarrow NO + O$

- **Rotational and translational modes are in equilibrium at a gas kinetic temperature**
- **Single vibrational quantum change processes dominate at low temperatures involved**
- **Significant body of theory and experimental validation data for the rates used**
- **Master equation coupled to Boltzmann equation for EEDF, species concentrations equations**
- **Nonequilibrium air plasma chemistry, excited electronic states kinetics are included**
- **Model validated using CARS $T_v(N_2)$ measurements in plane-to-plane nsec pulse discharge**

Montello et al, AIAA Paper 2012-3180

Air, “high current” regime: number of vibrational quanta per molecule vs. time





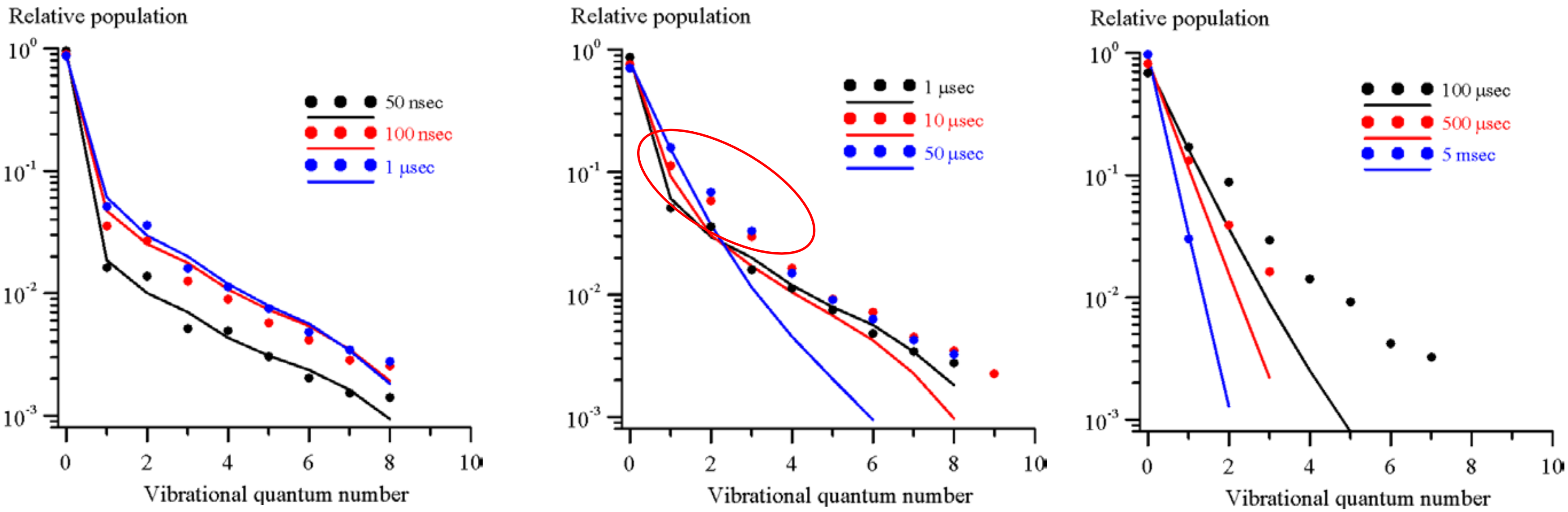
Average number of vibrational quanta per molecule (N_{quanta}):

$$N_{quanta} = \sum_{v=0}^{9} v f_v$$

- **Significant increase of number of vib. quanta per molecule <u>after the pulse</u> (a factor of 2)**
- **At variance with the model, which predicts N_{quanta}=const after the pulse (V-V exchange conserves quanta)**

Air, “high current” regime: $N_2(X,v)$ vibrational level populations vs. time

Nonequilibrium Thermodynamics Laboratories



The VDF evolution can be divided into 3 phases:

(1) Initial appearance and growth of all vibrational levels observed (Δt ~ 100 nsec – 1 μsec)

(2) Steady growth of low vibrational levels (v ~1-3) observed, while higher levels remain nearly constant (Δt ~ 1 μsec – 100 μsec)

(3) Vibrational energy decay: V-T relaxation (by O atoms) and diffusion (Δt ~ 100 μsec – 10 msec)

Air: time evolution of $N_2(v=0\text{-}3)$

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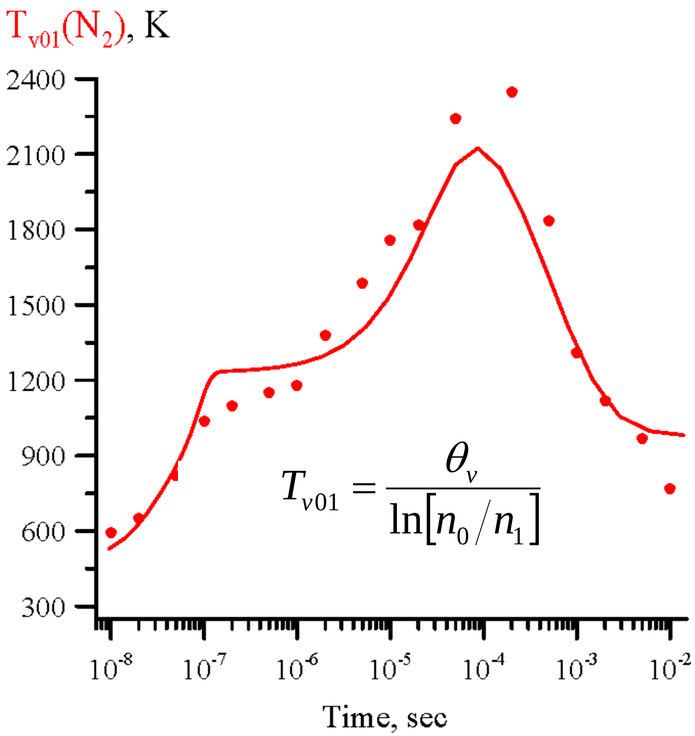

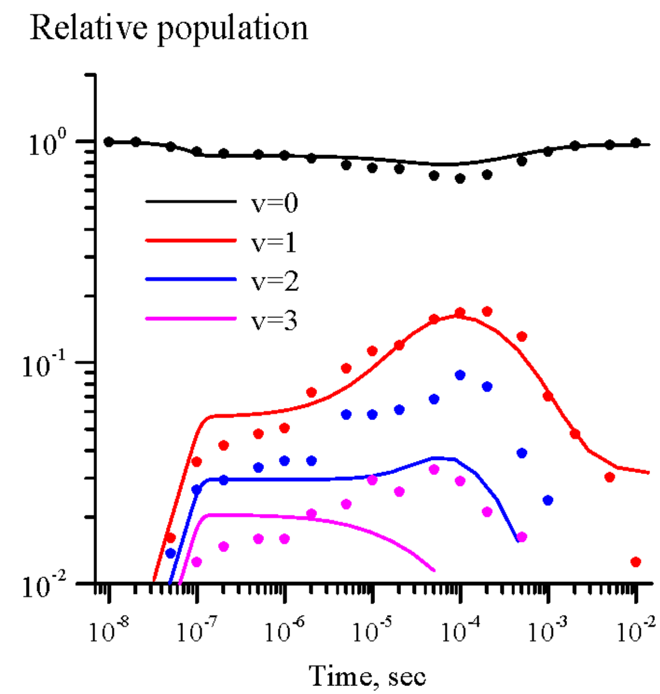

- **$T_{v01}(N_2)$ rise is primarily due to N_2–N_2 V-V exchange during relaxation: v=0, w → v=1, w-1**
- **Master equation model captures early VDF dynamics well: vibrational excitation by electron impact is modeled accurately**
- **As time evolves, v=0, 1 level populations are well predicted by model; higher level populations (v=2,3) are significantly underestimated. $T_{v01}(N_2)$ is not a good metric.**

Results in nitrogen are even more dramatic!

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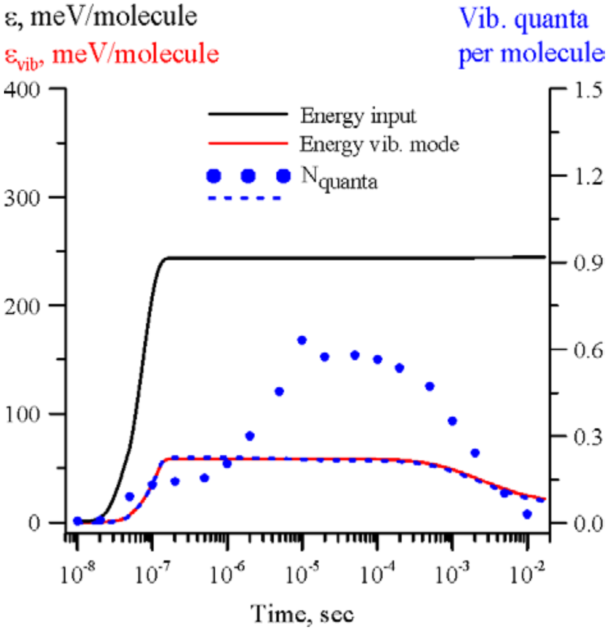
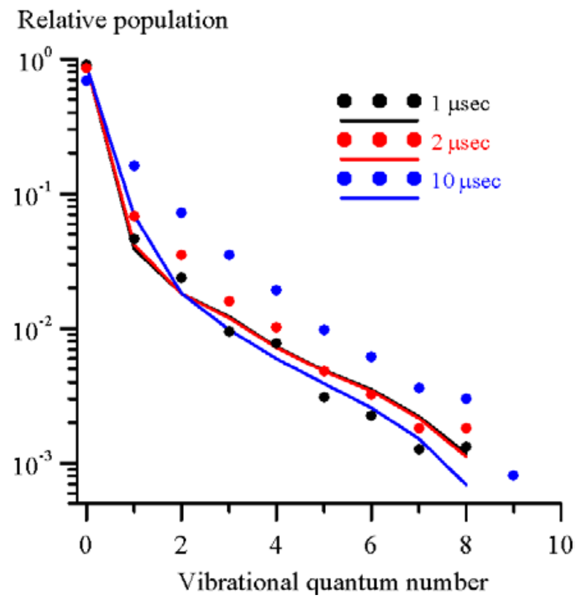
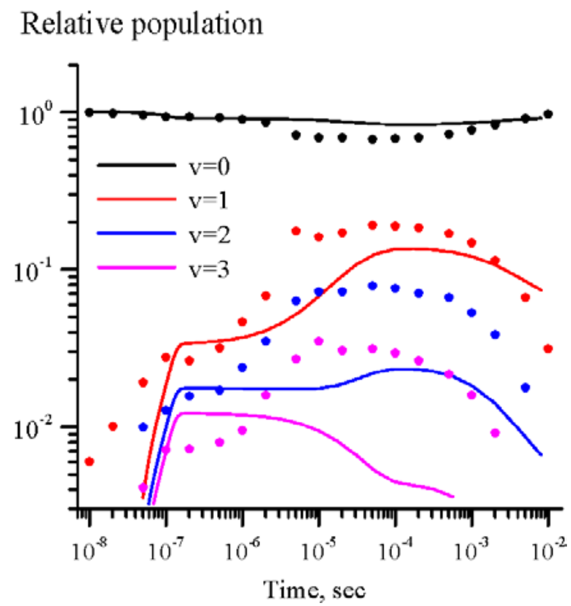
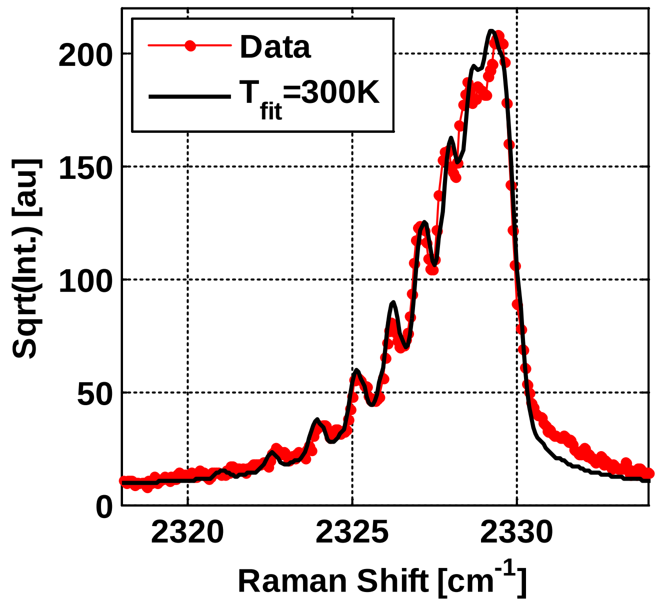




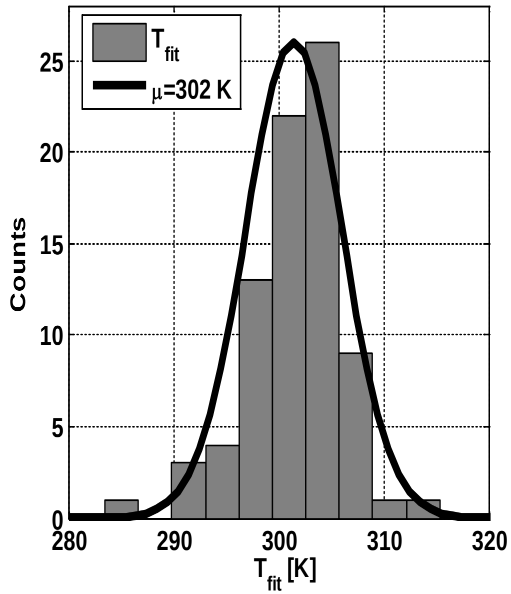


- N_{quanta} rise after the pulse by a factor of 4
- N_2(v=1-8) rise after the pulse by up to a factor of 5
- Not reproduced by the model

Rotational Temperature Measurements



100-shot accumulation spectrum in “cold” 100 torr air, with Sandia CARSFIT best fit synthetic spectrum.

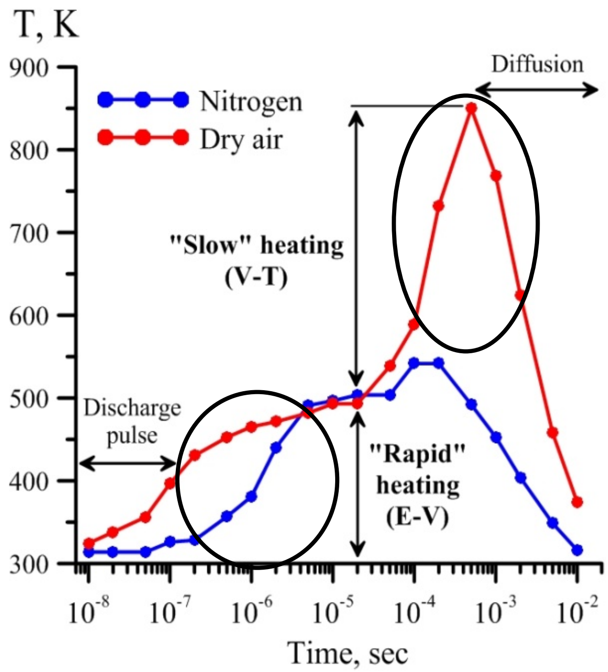


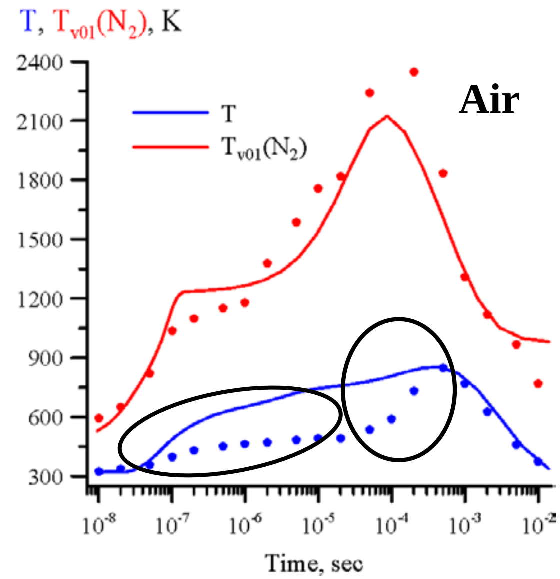
Histogram plot from measuring and fitting 80 such spectra.

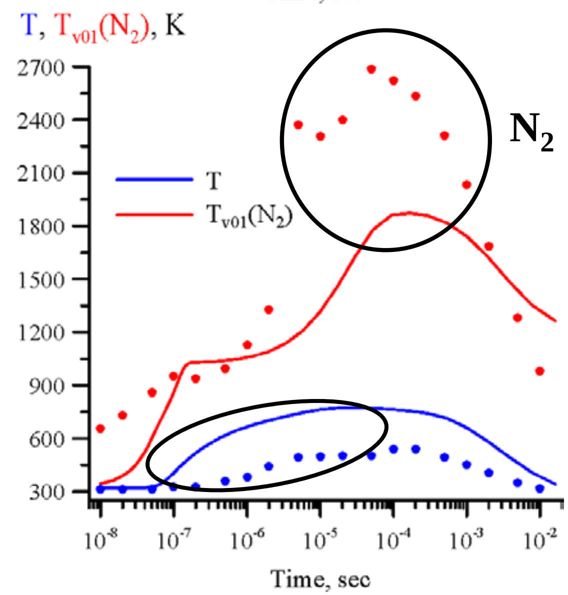
95% confidence interval ~ ± 9 K.

Rotational Temperature Results, Energy Balance

Nonequilibrium Thermodynamics Laboratories







- **Both in N_2 and air, model overpredicts "rapid" heating ,**
  likely N_2(A,B,C,a) + M → N_2(X,v) + M (E-V processes)
- **Energy stored in low N_2(X,v) underpredicted, especially in N_2**
- **Results suggest additional energy transfer into N_2(X,v) after the pulse: E-V processes ?**
- **In air, also model underpredicts "slow" heating (absent in N_2),**
  likely V-T relaxation by O: N_2(X,v) + O → N_2(X,v-1) + O

Effect of electronic-to-vibrational (E-V) energy coupling on energy balance in nsec pulse filament discharge in N_2

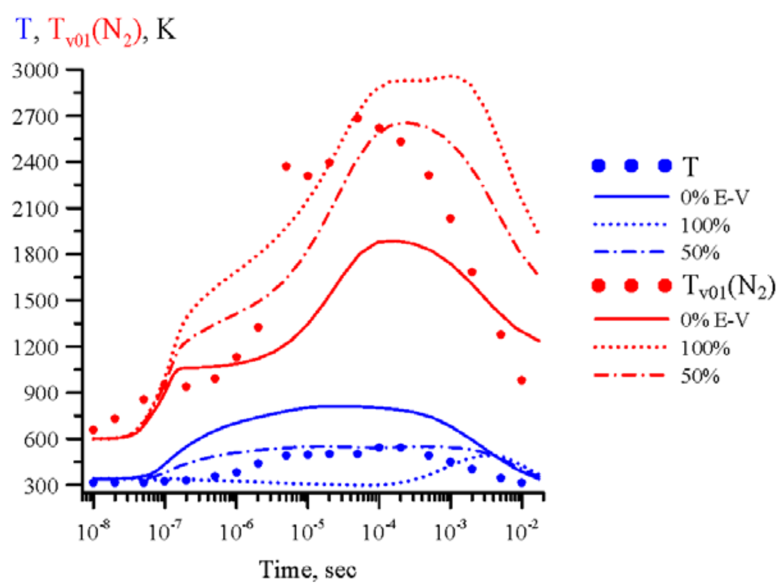
Nonequilibrium Thermodynamics Laboratories

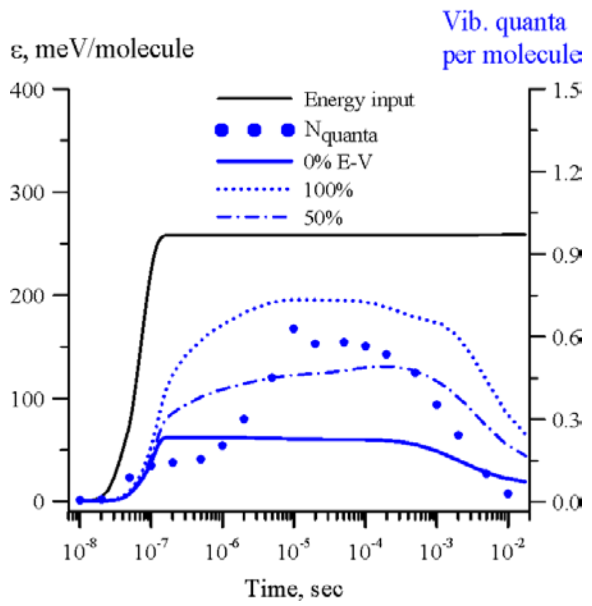
$$N_2(C)+N_2(X)\rightarrow N_2(B)+N_2(X,v)$$
$$N_2(A)+N_2(A)\rightarrow N_2(B,C)+N_2(X,v)$$

$$k(\rightarrow v)=k(T)\frac{\exp(-\alpha v)}{\alpha}$$

$$\bar{\varepsilon}_{vib}=\frac{\sum_v \varepsilon(v)k(\rightarrow v)}{k(T)}$$

α – adjustable parameter controlling percentage of energy defect into vibrational mode, $\bar{\varepsilon}_{vib}$





<u>Solid lines:</u> no energy into $N_2(X,v)$ during $N_2(A,B,C,a)$ quenching

<u>Dashed and Dash-dotted lines:</u> 100% or 50% of energydefect into $N_2(X,v)$

50% energy defect into $N_2(X,v)$ during N_2^* quenching: <u>better agreement</u> between measured and predicted $N_2(v)$

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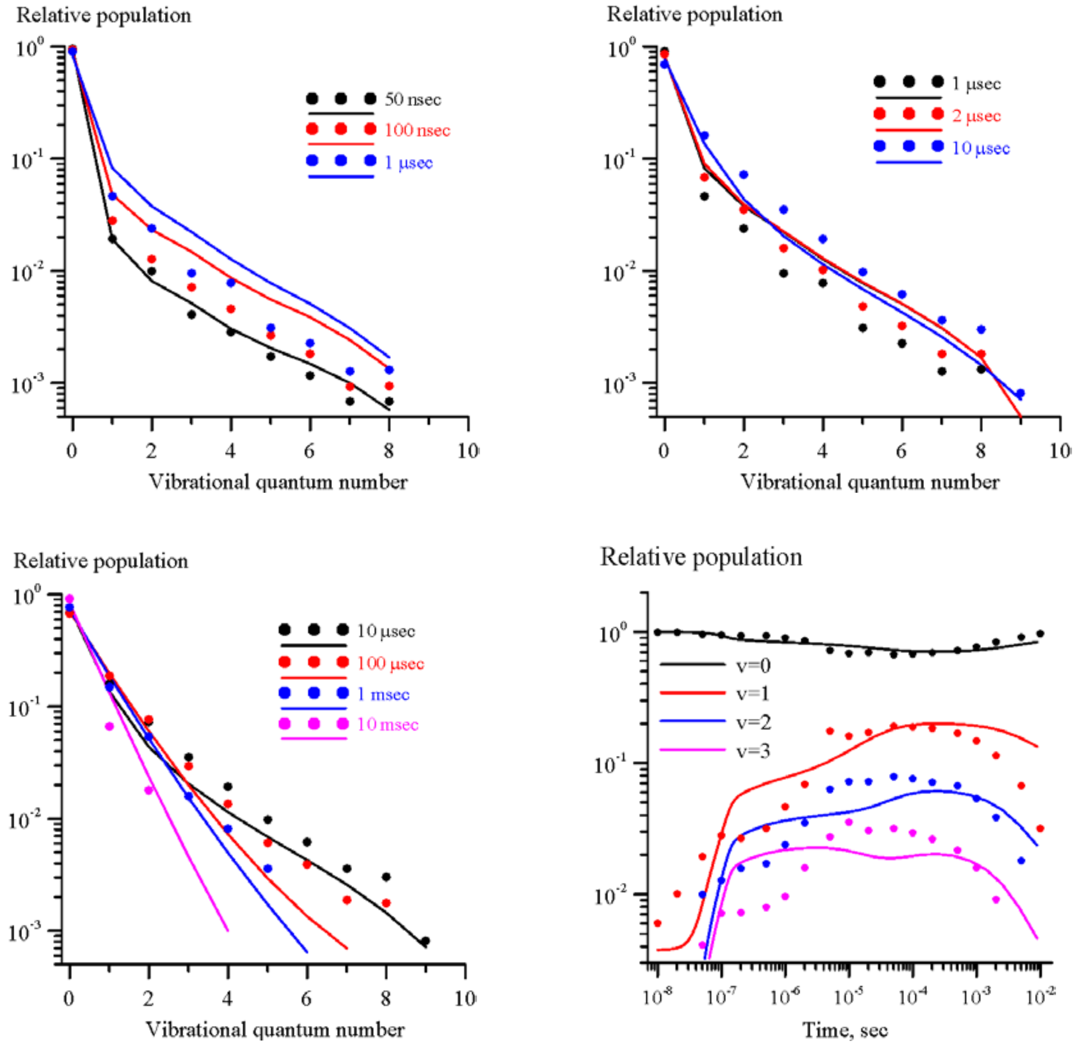
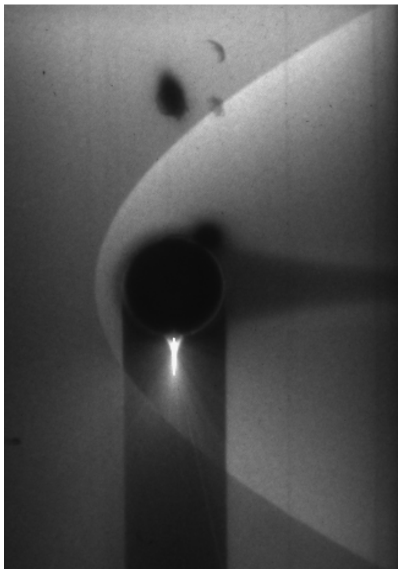
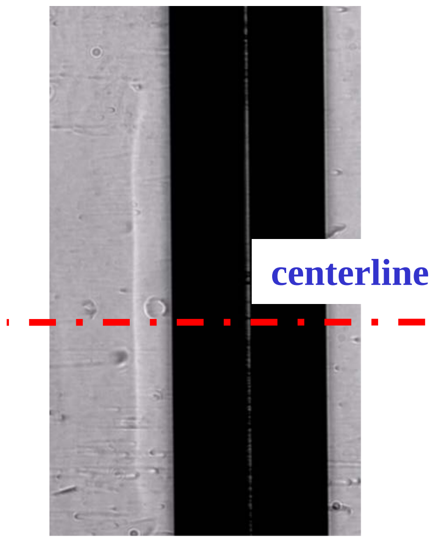


VDF dynamics at short time delays (Δt ~ 0.1 – 1 µsec) is still not reproduced well

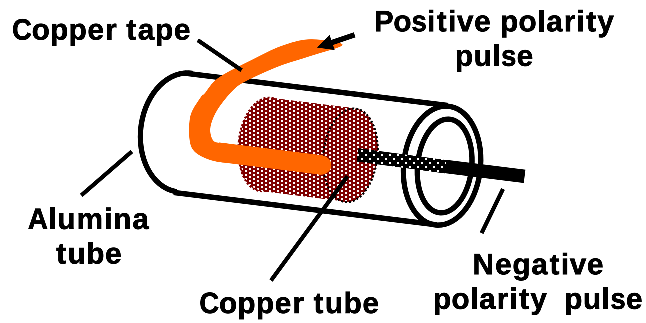
Here is why these kinetics are important:
NS DBD actuator on a cylinder model in a Mach 5 flow



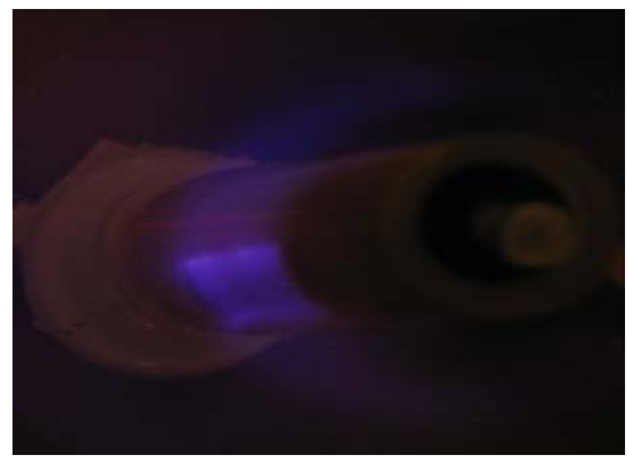
NO PLIF image
Nitrogen, P_0=0.5 atm



Schlieren image
Top view
5 mm diameter model
Stand-off distance 1.2 mm



Cylinder model / NS DBD plasma actuator
- Immersed electrode inside 6 mm quartz tube
- Exposed electrode: 1-3 mm wide copper strip

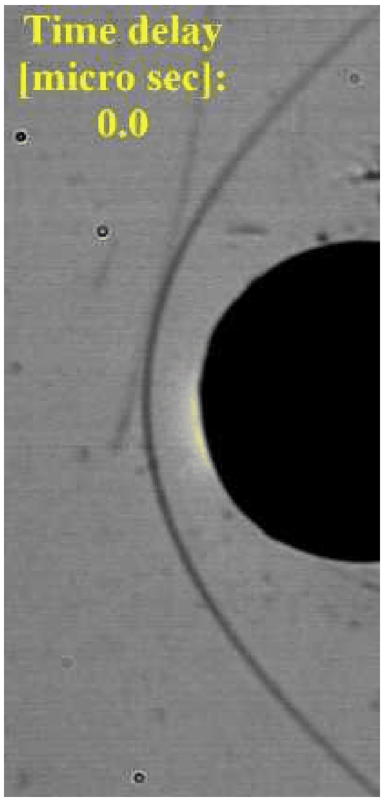


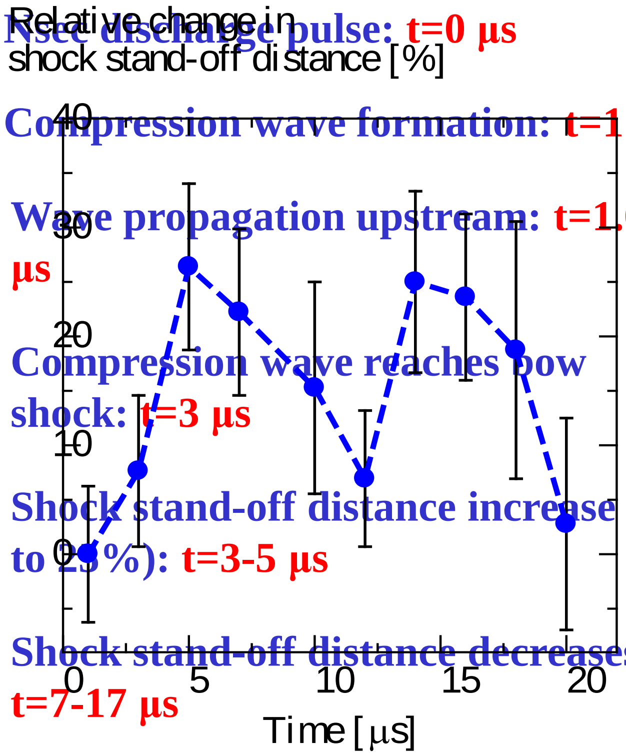
Plasma span ~1 cm, test section width 4 cm
T_R=340±30 K, ΔT=50 K (N_2 emission spectra)

Nishihara et al, Phys. Fluids, 2011

Phase-locked schlieren images of bow shock perturbations
Air, P_0=370 torr

Nonequilibrium Thermodynamics Laboratories





- Nsec discharge pulse: t=0 µs
- Compression wave formation: t=1 µs
- Wave propagation upstream: t=1.0-2.5 µs
- Compression wave reaches bow shock: t=3 µs
- Shock stand-off distance increase (up to 25%): t=3-5 µs
- Shock stand-off distance decreases: t=7-17 µs

Phase-locked schlieren signal

Difference from baseline (taken at t=0)

Repetitive shock perturbation if pulses repeated every 10 µs (at 100 kHz)

Nishihara et al, Phys. Fluids, 2011

Summary

Psec, broadband CARS:

- **Significant new advance in characterization of nonequlibrium flows: T_{rot} and $N_2(X,v)$ with high spatial (~50 x 500 μm) and time (~ 1 nsec) resolution**
- **Detailed new insight into kinetics of vibrational and electronic energy transfer, coupling to flow field**

Experiments in Mach 5 nonequilibrium wind tunnel:

- **Steady-state, vibrationally nonequilibrium flows generated in stable, high-pressure nsec pulser / DC sustainer discharge in plenum**
- **CARS measurements of $T_v(N_2)$, $N_2(X,v)$, and T_{rot} in nozzle plenum, with and without adding vibrational energy relaxers**
- **Spatially resolved CARS measurements of $T_v(N_2)$ in Mach 5 free stream flow and in bow shock layer; no detectable relaxation in the shock layer**

Experiments in single pulse, point-to-point (high power loading) nsec discharge:

- **Significant energy loading (up to ~0.3 eV/molecule/pulse)**
- **Spatially- and time-resolved N_2 VDF and T_{rot} indicates "feeding" of N_2 vibrations, e.g. $N_2(A) + N_2(A) \rightarrow N_2(C) + N_2(X,v)$, ~10-100 μs AFTER ~100 ns discharge pulse**

Future work

- **Elucidate mechanisms of coupling between microscopic molecular energy transfer and macroscopic flow parameters**

  (a) “rapid” (sub-acoustic time scale) heating → compression wave formation

  (b) vibrational energy transfer from “storage” to “relaxer” species, their subsequent relaxation → modulation of energy fluctuations spectra ?

- **Coupling between “rapid” heating kinetics and amplitude of compression waves: straightforward mechanism for high-amplitude, high bandwidth flow perturbations**

- **Possible coupling between rapid N_2-CO_2 V-V energy transfer / CO_2 V-T relaxation and amplification of acoustic perturbations (reverse of ultrasound absorption in molecular gases). Can this process be used to trigger flow acoustic instabilities?**

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