Picosecond CARS Measurements of Vibrational Distribution Function in a Nonequilibrium Mach 5 Wind Tunnel

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Abstract: Picosecond Coherent Anti-Stokes Raman Scattering (CARS) spectroscopy is used for measurement of nitrogen Vibrational Distribution Function (VDF) in the plenum of a highly nonequilibrium Mach 5 flow. First level nitrogen vibrational temperatures (T_v(N2)) of the order of 2000 K are achieved in the 300 Torr non-self-sustained plasma wind tunnel plenum. Downstream injection of CO2, NO, and H2 results in vibrational relaxation, demonstrating the ability to further tailor the vibrational energy content of the flow.

Keywords: Vibrational Distribution Function, Psec CARS, nsec pulsed discharge

1. Introduction

The ability to tailor non-equilibrium hypersonic flows by control of the loading of internal degrees of freedom (vibrational and electronic states), as well as dissociation and ionization fraction, is a topic of much current interest. For example, turbulent transition delay in a Mach 5 flow over a 5° cone by means of injection of carbon dioxide into nitrogen or air flow has been recently demonstrated [1]. Another example is the relaxation of energy stored in internal degrees of freedom of molecules behind a bow shock, which may significantly increase shock stand-off distance. Finally, short-pulse electric discharges which efficiently load electronic energy levels of nitrogen and oxygen in air are currently being explored as a means of hypersonic flow control, by producing repetitive localized pressure perturbations in the flow [2].

In this paper we provide new experimental data characterizing vibrational energy loading of molecular nitrogen in a hypersonic non-equilibrium flow vibrationally excited by a non-self-sustained electric discharge in the plenum of a Mach 5 wind tunnel. Temporally and spatially resolved vibrational level populations of nitrogen are measured using picosecond Coherent Anti-Stokes Raman Scattering (CARS). It is also shown that injection of gases such as CO2, NO, and H2 downstream of the discharge section, results in partial vibrational relaxation of the nitrogen excited in the discharge.

2. Experimental

The experiments in the current study were conducted in the plenum of a Mach 5 nonequilibrium flow tunnel, previously described in great detail [3,4]. Briefly, the lab scale blow-down facility operates at plenum pressures in the range of 0.25 – 1 atm, and has a steady-state run time of 5-10 seconds. The plenum section houses two fully overlapping electric discharges, (a) repetitively pulsed (up to 100 kHz), high peak voltage (up to 30 kV), short pulse duration (5 nsec) diffuse electric discharge and (b) transverse DC (5 kV, 2 A supply) sustainer discharge. The 4 cm x 4 cm copper plate pulser electrodes are flush mounted in the top and bottom walls of the nozzle plenum, separated by 0.5 cm, while the 4 cm x 1 cm copper plate sustainer electrodes are placed along the side walls of the pulsers discharge section, with 3.0 cm between them. This provides the ability to generate stable nonequilibrium plasmas at high pressures and high internal mode energy loading, while maintaining low translational-rotational temperatures in the range of T=350-400 K (as previously measured by N2 emission UV/visible spectroscopy [3,4]).

The high-frequency high-voltage pulser produces pulse-periodic, spatially uniform volume ionization in the discharge which does not have time to fully decay between pulses. The DC sustainer discharge, kept intentionally below breakdown threshold, couples significantly more energy to the ionized flow than the nanosecond discharge. In the
present experiment, the pulsed discharge is operated for 0.6 seconds, and the DC discharge for 0.5 seconds. The estimated reduced electric fields in the two discharges are significantly different, \((E/N)_{peak} \sim 300\) Td in the nanosecond pulsed discharge and \(E/N \sim 10\) Td in the DC discharge \((1\) Td = \(10^{-17}\) V·cm²). At these conditions, a significant fraction of input power in the pulsed discharge is spent on electronic excitation, dissociation, and ionization of nitrogen, while nearly all input power in the DC discharge (up to \(\sim 80-90\%\) [5]) is stored in the vibrational energy mode of nitrogen. Due to the very long \(N_2\) vibrational relaxation time at near room temperature, \(\sim 1\) atm·sec [6], this approach can create essentially vibrationally frozen nitrogen and air flows in the supersonic test section, with vibrational temperature \((T_v)\) greatly exceeding the translational / rotational mode temperature.

Downstream of the discharge section, the gas flows through a choked flow injector in which gases inducing vibrational relaxation are injected. The measurement location is 9 cm downstream from the gas injection, but upstream of the supersonic nozzle throat.

For direct measurement of vibrational energy loading provided by the pulser-sustainer discharge, the CARS spectroscopic technique has been employed. CARS is a four wave mixing technique that has been used extensively for thermometry of combustion and other gas phase reacting and non-reacting flows [7,8]. For this work, collinear phase-matched, psec vibrational CARS has been utilized. Through the use of a broadband psec Stokes laser source, patterned after that of Roy, et al [9], multiple vibrational levels can be interrogated simultaneously, whereby the Vibrational Distribution Functions (VDFs) of nitrogen can be obtained. More details of the optical diagnostic system can be found in Ref. [10].

3. Results and Discussion

Figure 1 shows a typical single laser shot psec CARS spectrum taken with the nanosecond pulser operating as described above and with the DC sustainer discharge off, plotted on a semilog scale. The \(v = 0\) and \(v = 1\) peaks are both clearly visible, indicating some vibrational excitation by operation of the pulser alone. The broad, low amplitude, non-resonant background is also clearly evident. Fig. 2 shows a typical single shot spectrum for the pulser-sustainer discharge in operation, with 4.5 kV DC voltage applied. As can be seen, the vibrational excitation for this case is much more significant than for the pulser alone, with vibrational levels \(v = 0, 1, 2, 3\) distinguishable, and higher levels possibly populated.

The spectral data can be reduced to yield the \(N_2\) VDF, the process of which is described in greater detail in Ref. [10]. The \(v=0\) and \(v=1\) number densities \((n_0\) and \(n_1)\) are then used to determine the “first level” nitrogen vibrational temperature, \(T_v(N_2)\), as follows:

\[
T_v(N_2) = \frac{\theta_{(v=1)}}{\ln[n_0/n_1]}
\]

where \(\theta_{(v=1)} = 3353\) K is the energy difference between vibrational levels \(v=1\) and \(v=0\) in the units of temperature. As the figures indicate, the pulser
operating alone yields $T_v(N_2)$ of 1100 K, while the pulser-sustainer discharge, with 4.5 kV DC, yields $T_v(N_2)$ of 2150 K. Uncertainty is estimated from the typical standard deviations seen in Fig. 3 below, which account for both measurement uncertainty and discharge variations.

With the laser timing synchronized to the pulser triggering system, the delay was modified in 10 msec intervals to allow measurement of $T_v(N_2)$ throughout the 0.6 sec discharge run time. Four measurements were taken at each time, and the results (mean +/- standard deviation) are shown in Fig. 3, with the DC sustainer current profile superimposed. These measurements were taken at 300 Torr plenum pressure and 3.5 kV DC. The figure illustrates the typical DC ramp time ~0.1-0.2 sec, due to capacitor charging in the power supply. As the figure shows, the CARS system sensitivity threshold is approximately 800-1000 K, which corresponds to the pulser alone condition, observed initially and after the DC supply is powered off. Work is currently underway using an alternate CARS alignment strategy which should yield greater signal to noise and lower the system measurement threshold. Taking the data from $t = 200$ msec to $t = 590$ msec as steady-state, this configuration yields an average $T_v(N_2) = 1671$ K with average 95% confidence interval of 25 K. Due to the very slow vibration-translation relaxation in $N_2$, ~1 sec for these conditions, these results represent a quasi-steady-state, time-resolved vibrational temperature reached in the pulser-sustainer discharge. Spectra taken after the pulser is turned off show no detectable $v = 1$ excitation, which seems to indicate this is a purely non-thermal effect.

Figure 4 shows $T_v(N_2)$ data as a function of DC $E/n$ for three different pressures, assuming 300 K translational temperature. As is well known [5], $E/n$ controls the average electron energy and therefore the input energy partition among different electron impact excitation channels (vibrational and electronic excitation, dissociation, ionization, etc). Each data point in the plot represents the mean and standard deviation from the 4-5 single shot spectra taken at each condition. As can be seen, increasing the DC sustainer voltage, i.e. the energy loading in the discharge, results in higher vibrational excitation of nitrogen in the flow, as expected. As the figure shows all three pressures behave very similarly for $E/n < 10$ Td. For the 200 Torr case, lower vibrational excitation is observed beyond this region than that observed for the other two pressures, which remain in good agreement with each other. The lower $T_v(N_2)$ measured for the 200 Torr case could result from discharge spatial non-uniformity (visible emission is seen to be most intense near the DC electrodes along the side walls), or from a decrease in power loading per molecule.

![Figure 3. Temporal evolution of $T_v(N_2)$ shown with sustainer discharge current profile, 3.5 kV DC, 300 Torr $N_2$.](image)

![Figure 4. Vibrational temperature vs. estimated reduced electric field in the sustainer discharge (1 Td = $10^{-17}$ V·cm²).](image)

The final set of experimental data, shown in Fig. 5, was obtained with injection of gases inducing vibrational relaxation of the discharge-excited nitrogen. For this work, injection of $O_2$, $H_2$, NO, CO$_2$ was performed. These species were chosen because the rates of nitrogen vibrational relaxation in these four mixtures vary by several orders of magnitude, seen in Table 1 below. To summarize
the results briefly, the injection of oxygen, up to 17% (nearly “synthetic air”) results in negligible reduction in $T_v(N_2)$, due to an extremely slow energy transfer rate, while CO$_2$ injection results in drastic vibrational energy reduction. The results from nitric oxide and hydrogen injection lie between these two extremes, with NO behaving as expected, while the H$_2$ injection appears to have more effect than the rate coefficient would suggest. This is not surprising however, as this rate information was extracted from shock tube measurements, and could very likely not be valid for this low translational temperature regime. These measurements demonstrate the feasibility of generating high-pressure, low-temperature vibrationally excited flows of nitrogen and synthetic air at steady state, as well as feasibility of tailoring the vibrational temperature of these nonequilibrium flows using injection of efficient V-T and V-V relaxer species downstream of the discharge.

<table>
<thead>
<tr>
<th>Energy Exchange mechanism</th>
<th>Rate coeff. [cm$^3$/sec], 300 K</th>
<th>Injection pressure [Torr]</th>
<th>Char. Lifetime</th>
</tr>
</thead>
<tbody>
<tr>
<td>O$_2$ (v-v)</td>
<td>$7 \times 10^{-17}$ [11]</td>
<td>50</td>
<td>10 msec</td>
</tr>
<tr>
<td>CO$_2$ (v-v)</td>
<td>$6 \times 10^{-15}$ [11]</td>
<td>5</td>
<td>10 usec</td>
</tr>
<tr>
<td>NO (v-v)</td>
<td>$1.4 \times 10^{-15}$ [12]</td>
<td>5</td>
<td>4 usec</td>
</tr>
<tr>
<td>H$_2$ (v-t)</td>
<td>$1 \times 10^{-16}$ [13]</td>
<td>5</td>
<td>60 msec</td>
</tr>
</tbody>
</table>

Table 1. Relaxation rate coefficients and characteristic lifetimes for various relaxant species injection partial pressures.

4. Conclusions

Picosecond CARS spectroscopy is used for measurement of nitrogen Vibrational Distribution Functions in the plenum of a highly nonequilibrium Mach 5 flow, created by a pulser-sustainer discharge. First level vibrational temperatures of ~2000 K are achieved at pressures as high as 370 Torr. Downstream injection of CO$_2$, NO, and H$_2$ results in vibrational relaxation, demonstrating the ability to further tailor the vibrational energy content of the flow.

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References