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Rotation-vibration non-equilibrium measurement using pure rotational fs/ps CARS coherence beating

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Abstract: Rotation-vibration non-equilibrium plays a key role in the chemistry and thermalization in low-temperature plasmas as well as thermal loading of hypersonic vehicles. Here, we report the development of a simple and sensitive two beam hybrid femtosecond/picosecond pure rotational coherent anti-Stokes Raman scattering (fs/ps CARS) approach to simultaneously measure the rotational and vibrational temperature of N_2. This approach takes advantage of probe delay specific interferences between ground state and vibrationally excited N_2 molecules to intentionally induce so-called "coherence beating," that lead to apparent non-Boltzmann distributions in the pure rotational fs/ps CARS spectra. These distortions in the spectra enable simultaneous inference of both the rotational and vibrational temperatures. The coherence beating effects were observed in single shot fs/ps CARS measurements of a 75 Torr N_2 DC glow discharge and were successfully modeled for rotational and vibrational temperature extraction. Rotational temperatures around 400 K and vibrational temperatures from 1000 K to 4500 K were measured. It is shown that this method can be more sensitive than a pure rotational fs/ps CARS approach using a spectrally narrow probe pulse. This method is general to most diatomic molecules and opens the possibility of high speed 1-D and 2-D simultaneous rotation-vibration non-equilibrium measurements in plasmas and hypersonic flows.

1. Introduction

Rotation-vibration non-equilibrium measurements are critical for understanding energy transfer and thermalization in low-temperature plasmas and hypersonic vehicles. In plasmas, energy transfer to molecules via collisions with electrons can induce significant vibrational non-equilibrium. Vibrationally-excited molecules are hypothesized to enhance chemical reactivity in plasma-catalysis [1], plasma-assisted combustion [2], and plasma CO_2 dissociation [3]. Furthermore, in hypersonic flows, shock-boundary layer interactions can induce rotation-vibration non-equilibrium which affects the thermal loading and aerodynamics of hypersonic vehicles [4,5]. Simultaneous measurements of both the rotational and vibrational temperature distributions are necessary to further our understanding in these systems.

Hybrid fs/ps coherent anti-Stokes Raman scattering (fs/ps CARS) is a powerful laser diagnostic capable of probing gas temperature and species concentrations on the picosecond time scale with tens of microns of spatial resolution [6–14]. A method for retrieving vibrational and

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rotational temperatures simultaneously from spatially-resolved 1-D rotational fs/ps CARS images was recently developed [13]. This method uses the red-shift of rotational energy levels of vibrationally-excited molecules to measure the vibrational temperature along with the rotational temperature. The key advantages of this approach were the two-beam configuration for straightforward 1-D imaging and the elimination of the need for a separate Stokes pulse generated from an optical parametric amplifier (OPA). However, in [13] a 20 Hz repetition rate picosecond laser with a narrow line-width (~0.23 cm^{-1}) was used to resolve the one-sided broadening induced by vibrationally-excited N_2. It would be preferable to use a probe pulse generated from the femtosecond laser and enable kHz data acquisition rates.

Narrow probe picosecond pulses for fs/ps CARS have been previously generated from femtosecond pulses using second harmonic bandwidth compression [10,11,15–19]. However, these probe pulses typically have 3 to 6 cm^{-1} bandwidths, while the energy spacing between adjacent vibrational levels in the pure rotational spectrum is approximately 1 cm^{-1} in N_2. Therefore, multiple Raman transitions from different vibrational levels will be coherently sampled within the probe pulse bandwidth and they can interfere with one another. This induces so-called "coherence beating" between these Raman transitions and distort the CARS spectrum as a function of probe delay. For example, in the rotational O_2 spectrum, coherence beating occurs due to the electronic triplet configuration of ground state O_2 which splits each rotational energy level into three closely spaced energy levels [20]. For certain probe delays, significant distortion of the CARS spectrum was observed, and accurate temperature evaluations were not possible without considering the electronic triplet state splitting. To mitigate coherence beating effects, dual-pump fs/ps CARS was developed for rotation-vibration non-equilibrium measurements in [10]. Recent simultaneous rotation-vibration non-equilibrium measurements in Mach 18 flows were performed using a similar method [14]. However, such measurements require multiple cameras and an OPA to access the vibrational CARS spectrum. This adds complexity and reduces the pulse energy available for generating a CARS signal due to losses in the OPA. It would be desirable to retain two-beam CARS for rotation-vibration non-equilibrium measurements like in [13].

In this study, we propose to develop a new diagnostic method which uses a picosecond probe pulse generated by second harmonic bandwidth compression (SHBC) directly from the fs laser and utilizes the signatures of the time-domain coherence beating to simultaneously infer the rotational and vibrational temperature from the pure rotational fs/ps CARS spectrum. The sensitivity of this approach was compared to the frequency-domain method previously developed in [13]. A N_2 DC glow discharge was used as a test bed for experimentally demonstrating this technique. Vibrational temperatures ranging from 1300 K to 4000 K were measured on a single shot basis using this method.

2. Experimental Methods

The detailed theory and principles behind fs/ps CARS frequency and time-domain modelling can be found in [8,13,21] and only a brief overview will be given here. Hybrid fs/ps CARS takes advantage of the broadband nature of femtosecond laser pulses to probe multiple Raman transitions simultaneously, while the non-resonant background is avoided with a time delayed spectrally-narrow probe pulse. In this study, two-beam phase matching is used where the both the pump and Stokes photons come from a single fs laser source [22]. The principle behind detecting vibrational and rotational temperatures simultaneously from the rotational is given by equations (1-3):

$$\boldsymbol{F(v,J) = B_v(J(J+1)) - D_v\left(J^2(J+1)^2\right)} \qquad \textbf{(1)}$$

$$B_v = B_e - \alpha\left(v + \frac{1}{2}\right) + \gamma\left(v + \frac{1}{2}\right)^2 \quad (2)$$

$$D_v = D_e + \beta\left(v + \frac{1}{2}\right) \quad (3)$$

where F is the rotational energy of an N_2 molecule in a state (v, J) with v and J being, respectively, the vibrational and rotational energy levels, B_v and D_v the vibrational energy level-dependent rotational and centrifugal constants, B_e and D_e are the equilibrium rotational and centrifugal constants, and α, β, and γ are rotation-vibration coupling constants. From equations (1) to (3), the rotational energy levels exhibit a red-shift with increasing vibrational energy due to the decrease in B_v and increase in D_v. This vibrationally-induced shift can be on the order of 1 cm^{-1}. For molecules in rotation-vibration non-equilibrium, molecules in the excited vibrational states will appear at these shifted wavelengths. Such shifts have been detected before using spontaneous Raman scattering and nanosecond pulse-width CARS [23,24]. The coherence beating induced by interference between the neighboring vibrational levels allows for inference of the vibrational temperature in the pure rotational CARS spectrum. This approach is similar to [25], where an optimal probe delay for vibrational Q-branch thermometry of N_2 was found due to the interferences between neighboring rotational lines.

The fs/ps CARS setup is shown in Fig. 1. The pump/Stokes photons were supplied by a femtosecond Ti:Sapphire regenerative amplifier (Coherent Astrella), and a 9 ps probe pulse was generated using a second harmonic bandwidth compression (SHBC) design similar to [26]. A 1500 gr/mm diffraction grating with 90% diffraction efficiency (Spectrogon) was used as the dispersive element to stretch the femtosecond pulse and the stretched beam was split via a 50/50 beam splitter (Thorlabs). Propagation through the beam splitter and reflection by the beam splitter produces two spatially chirped pulses that are mirror images of each other. In other words, they are oppositely chirped, as required for spectral compression via SHBC. A 250 mm focal length achromatic lens was used to collimate the dispersed light. Since the dispersion was only in one axis, a sheet was formed at the Fourier plane, where the nonlinear beta barium borate (BBO) crystal was placed. Temporal alignment of the split pulses was achieved through adjusting a micrometer translation stage and probe pulses with energies up to 100 μJ were generated. The femtosecond pulse and the picosecond pulse were spherically focused separately and directed to intersect at a 3-degree angle in a two-beam phase matching configuration. The spatial overlap was approximately 1 mm.

The time delay between the probe pulse and pump/Stokes pulse was set with a delay line on a motorized translation stage. Detection of the CARS signal was performed via an angle-tuned bandpass filter (Semrock), a Czerny-Turner spectrometer (Princeton Instruments), and an intensified charge coupled device (ICCD, Princeton Instruments). The plasma reactor was the same as in [13], but the electrodes have been swapped for a pin to plane discharge geometry with a pin anode and a 1 cm diameter cylindrical cathode separated by 7 mm. The anode was connected to a high voltage direct current (DC) power supply (Glassman) and a 100 kOhm ballast resistor. The voltage at the anode was 500 V and the potential drop across the resistor was 1500 V. The synchronization between the laser and the ICCD is achieved using a delay generator (Stanford Research Systems) triggered by the femtosecond laser. A constant flow of 600 SCCM of pure N_2 was maintained using a mass flow controller (MKS) and the pressure was kept constant with a downstream automatic control valve (MKS).

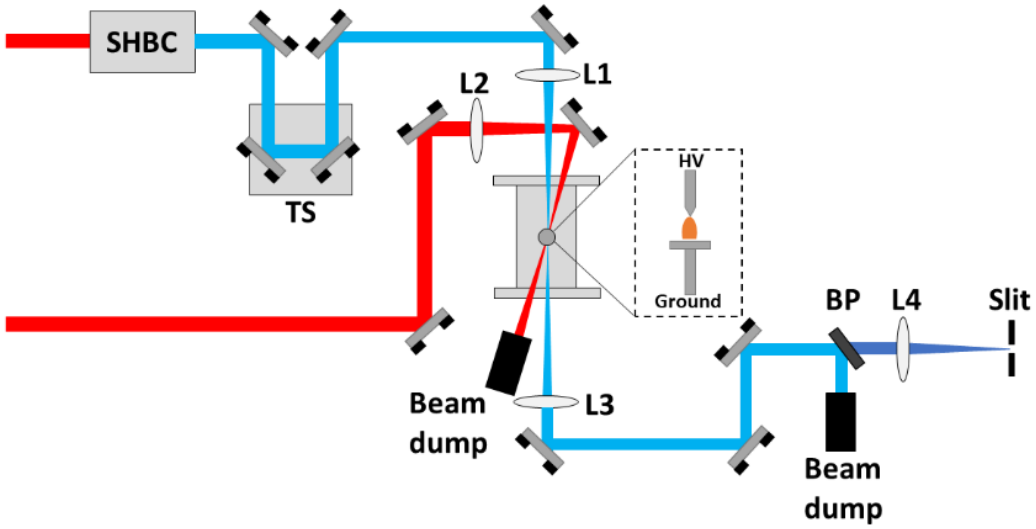

Figure 1. Experimental fs/ps CARS schematic. L1, L2, L3, L4: spherical plano-convex lenses with f = 400 mm, 400 mm, 400 mm, 75 mm. BP: angle-tuned bandpass filter

3. Results and Discussion

fs/ps CARS modeling results demonstrate the capability of using time-domain coherence beating for detecting rotation-vibration non-equilibrium. A 10-picosecond probe pulse was used in the simulations. In Fig. 2, the modelled spectrum of an equilibrium ($T_{vib} = T_{rot}$) 500 K spectrum is compared with a non-equilibrium spectrum with T_{rot} of 500 K and T_{vib} of 4000 K. A stark difference in both profiles can be seen. The equilibrium spectrum produces an apparent Boltzmann distribution of rotational states. However, the non-equilibrium spectrum is strongly distorted, due to the presence of coherence beating between vibrationally excited N_2 and the ground state N_2.

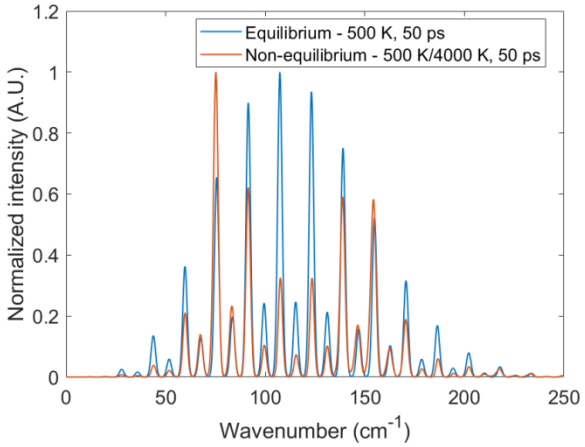

Figure 2. Modeling of the fs/ps pure rotational CARS N_2 spectra at a time-delay of 50 ps and pressure of 1 atm in rotation-vibration equilibrium and non-equilibrium.

This distortion is a function of the vibrational temperature since the beating will be stronger with more molecules in vibrationally excited states. This can be seen in Fig. 3 where the effects of

coherence beating becomes more severe with higher vibrational temperatures. This suggests that this effect can be leveraged as a sensitive and quantitative diagnostic for simultaneous rotational and vibrational temperature measurements. Furthermore, these intensity distortions are much larger than the one-sided broadening observed in [13]. Therefore, using the time-domain interferences can potentially be more sensitive than when using frequency domain detection. In Fig. 4, we compare the two approaches at a 50 ps delay and a vibrational temperature of 1000 K. If the frequency resolution of the measurement is limited by the probe bandwidth and not the spectrometer, then both the time and frequency domain techniques can measure this condition. However, typical grating spectrometers have instrument spectral broadening functions of ~ 0.5 to 1 cm^{-1}. In this case, the instrument broadening smears the adjacent peaks and limits the sensitivity of the measurement as shown in the Fig. 4. The equilibrium and non-equilibrium instrument broadened spectra are nearly indistinguishable at this vibrational temperature. For time-domain coherence beating, this is not a problem, and the theoretical sensitivity remains the same.

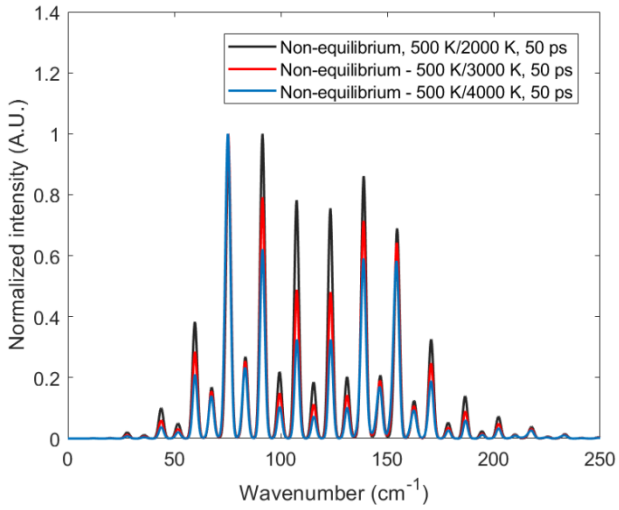

Figure 3. Variation of pure rotational N_2 fs/ps CARS spectrum with vibrational temperature at a probe delay of 50 ps and pressure of 1 atm.

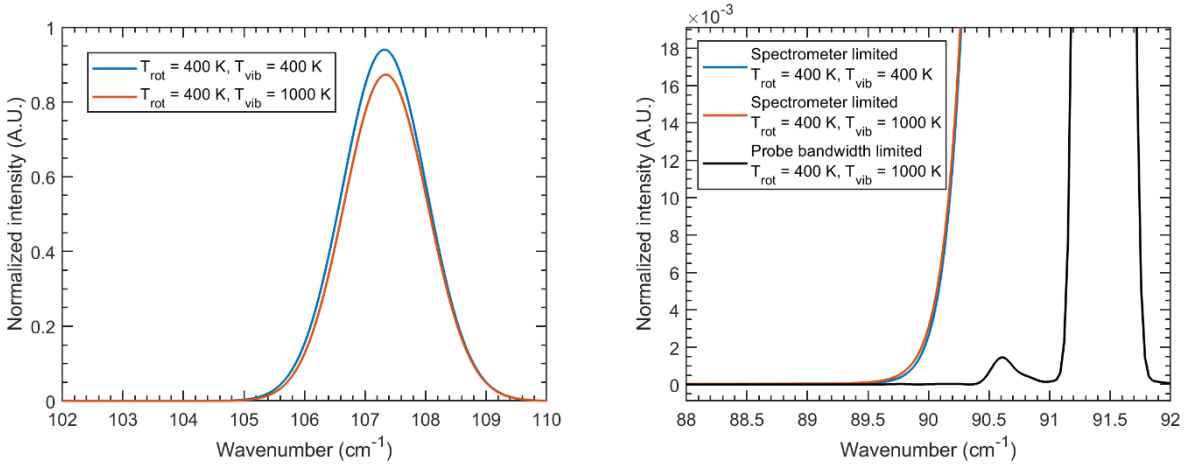

Figure 4. Comparison of the time-domain approach using a 9 ps probe (left) and the frequency domain using a 65 ps probe (right) for simultaneous detection of rotation-vibration non-equilibrium in the pure rotational CARS spectrum at T_{rot} = 400 K and T_{vib} = 1000 K.

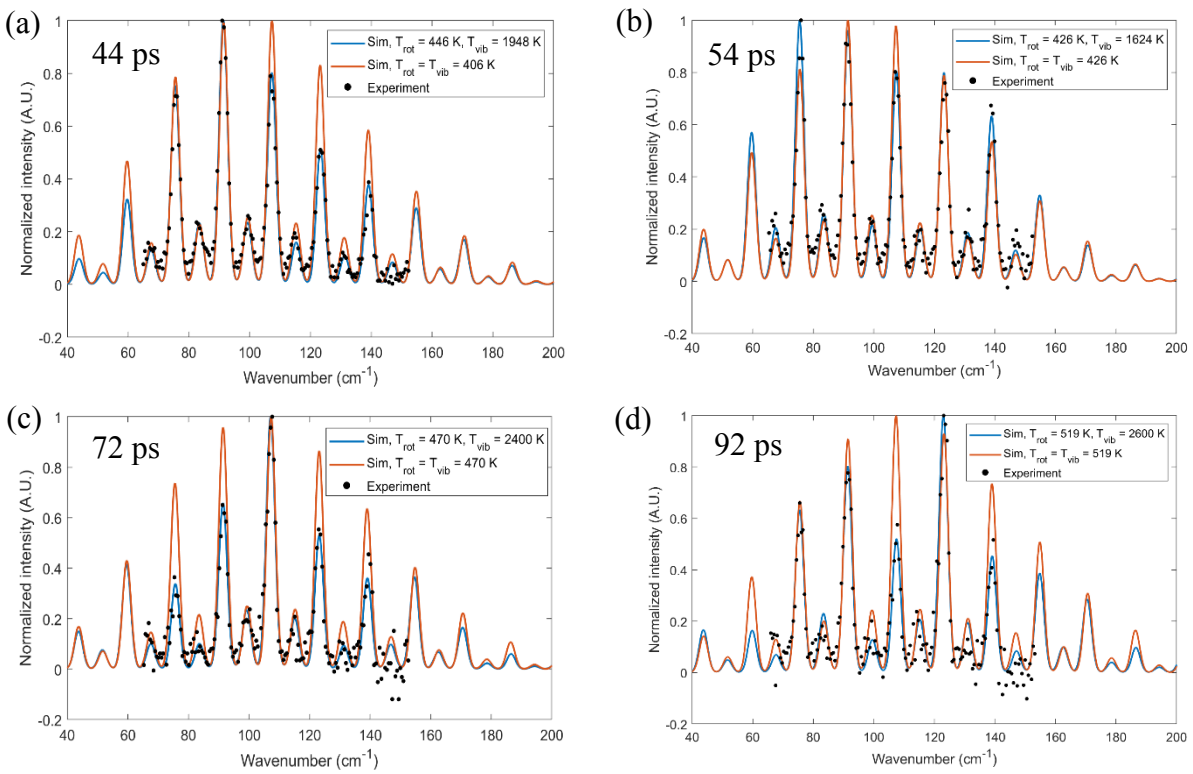

Figure 5. Measured single shot fs/ps CARS spectra of a N_2 glow discharge and corresponding simulated fits for rotation-vibration non-equilibrium at different probe delays. Spectra for the equilibrium conditions at the same evaluated rotational temperature were plotted for reference.

To experimentally validate the model simulations and demonstrate this technique, we measured fs/ps CARS spectra in a 75 Torr N_2 DC glow discharge plasma. Due to the low reduced electric field, efficient pumping of vibrational energy levels induced rotation-vibration non-equilibrium in this plasma. However, the DC glow discharge drifted in space, so single shot CARS measurements were necessary since averaging was not possible. Shown in Fig. 5 are the experimental data of this glow discharge at different probe pulse delays as well as the non-equilibrium fits. The effects of coherence beating on the spectra are immediately obvious from comparison of the data and the rotation-vibration equilibrium simulation. Varying the probe delays induced different beat patterns in the rotational spectrum and these coherence beating effects were successfully fitted by the CARS model. For the 92 ps delay, a clear deviation from a Boltzmann distribution of rotational states can be seen, while at 44 ps, the distortion is less obvious. This suggests that, like in [25], there should be an optimal probe delay to maximize the sensitivity and accuracy of the measurement. Such investigations will be pursued in the future.

Lastly, the time-dependent rotation-vibration non-equilibrium using a probe delay of 92 ps was measured in the discharge as shown in Fig. 6. Due to the non-stationary behavior of the discharge, both rotational and vibrational temperatures fluctuated as time progressed. Around 17 seconds, the vibrational temperature and rotational temperature both peaked at 4500 K and 600 K, respectively. This likely corresponded to the optimal overlap between the core of the positive column and the CARS probe volume.

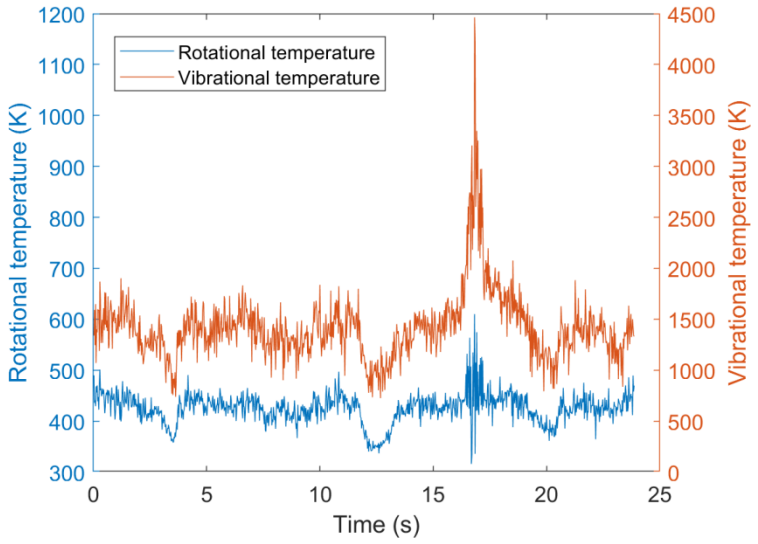

Figure 6. Time dependent rotation-vibration non-equilibrium measured in a 75 Torr N_2 DC glow discharge with a probe delay of 92 ps.

4. Conclusions

In this work, a new method to simultaneously measure rotational and vibrational temperatures of molecules using coherence beating of pure rotational hybrid fs/ps CARS was developed. The distortion of the pure rotational fs/ps CARS spectrum increases with the vibrational temperature, which indicates that both rotational and vibrational temperature can be directly retrieved from the pure rotational fs/ps CARS spectrum. Coherence beating between rotational lines of vibrationally excited and ground state N_2 was observed experimentally in a N_2 DC glow discharge. Furthermore, the feasibility of extraction of the rotational and vibrational temperatures from fitting the fs/ps CARS spectra distorted by the coherence beating was demonstrated. This technique was shown to be more sensitive than using a high frequency resolution probe for experiments limited by the spectrometer resolution. Moreover, the switch to measurement of intensity distortions in the spectra may enable the possibility of sensitive 2-D rotation-vibration non-equilibrium measurements using hyperspectral 2D-CARS [27,28] as well as high speed kHz two-beam fs/ps CARS detection of rotation-vibration non-equilibrium in plasmas [13] and hypersonic wind tunnels [11,14].

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