Nonlinear CARS measurement of nitrogen vibrational and rotational temperatures behind hypervelocity strong shock wave

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Abstract. The hypervelocity strong shock waves are generated, when the space vehicles reenter the atmosphere from space. Behind the shock wave radiative and non-equilibrium flow is generated in front of the surface of the space vehicle. Many studies have been reported to investigate the phenomena for the aerospace exploit and reentry. The research information and data on the high temperature flows have been available to the rational heatproof design of the space vehicles. Recent development of measurement techniques with laser systems and photo-electronics now enables us to investigate the hypervelocity phenomena with greatly advanced accuracy. In this research strong shock waves are generated in low-density gas to simulate the reentry range gas flow with a free-piston double-diaphragm shock tube, and CARS (Coherent Anti-stokes Raman Spectroscopy) measurement method is applied to the hypervelocity flows behind the shock waves, where spectral signals of high space/time resolution are acquired. The CARS system consists of YAG and dye lasers, a spectroscope, and a CCD camera system. We obtain the CARS signal spectrum data by this special time-resolving experiment, and the vibrational and rotational temperature of N₂ are determined by fitting between the experimental spectroscopic profile data and theoretically estimated spectroscopic data.

1. INTRODUCTION
With the development in aerospace science and technology, the opportunities of space explanation and reentry to the atmosphere have been increasing. When the spacecrafts reenter the atmosphere, the hypervelocity strong shock waves are generated in front of the vehicles. Behind the strong shock waves, the high temperature and non-equilibrium radiating flow are generated. This high temperature and non-equilibrium flow field is a very important subject to design the heat shields of the spacecrafts.

In previous study we simulated this hypervelocity strong shock wave with a free-piston double-diaphragm shock tube, and applied CARS system to the direct measurement of the vibrational/rotational temperatures of nitrogen molecule behind the radiating shock wave. This flow field can be regarded of a multiphase flow of vibrational and rotational exited gas molecules and ionaized plasma. In this study we obtain more CARS spectra data and measure the vibrational and rotational temperature of N₂ molecule in high temperature and radiating multiphase flow of plasma and air.

2. THEORETICAL CALCULATION OF CARS
The theory and experiment of CARS measurement have been developed extensively (Alan C.
Eckbreth., 1984; R. J. Hall., 1979; P. R. Régnier and P. E Taran., 1973), though they are restricted relatively slow gasdynamic phenomena from ms-orders as combustion experiments. According to the theory on CARS, incident YAG laser beams at frequencies $\omega_1$ (pump beam) and $\omega_2$ dye laser (Stokes beam) interact through the third order nonlinear electric susceptibility $\chi^{(3)}_{\text{CARS}}$ to produce a coherent radiation $I_3(\omega_3)$ at $\omega_3 = 2\omega_1 - \omega_2$ of the intensity written, as follows

$$I_3(\omega_3) = \frac{\omega_3^2}{n_1^2 n_2 n_3 c^4 \epsilon_0^2} \left| \chi^{(3)}_{\text{CARS}} \right|^2 I_1^2(\omega_1) I_2^2(\omega_2) \left( \sin \frac{\Delta k I}{2} \right)^2$$

(1)

$$\Delta k = 2k_1 - k_2 - k_3$$

(2)

where $n_1$, $n_2$ and $n_3$ are the refractive indexes at $\omega_1$, $\omega_2$, $\omega_3$, respectively; $c$ is the velocity of light; $l$ is interaction length; $k_1$, $k_2$ and $k_3$ are the wave vectors of the pump, Stokes and CARS beams, respectively; $\epsilon_0$ is the permittivity of free space. The CARS signal is enhanced on the condition of phase matching, $\Delta k = 0$. The incident laser beams, i.e. two pump beams ($k_1$) and a Stokes beam ($k_2$), are aligned in order to satisfy vector relation, where $\chi^{(3)}_{\text{CARS}}$ is shown by

$$\chi^{(3)}_{\text{CARS}} = \sum_j K_j \frac{\Gamma_j}{2\Delta \omega_j - i\Gamma_j} + \chi_w$$

(3)

Here the $j$ summation is over vibration-rotation transitions in the vicinity of $\omega_1 - \omega_2$; $\Gamma_j$ is the Raman line width (FWHM); the non-resonant term $\chi_w$ is a background contribution due to electrons and other remote resonances; and $K_j$ is shown to be related to the Raman cross-section

$$K_j = \frac{2n_c c^4}{n_2 h \omega_2^4} \frac{d\sigma}{d\Omega} \frac{N\Delta j}{\Omega_j}$$

(4)

where $N$ is the number density of the Raman active molecule; $\Delta j$ is the population difference between the upper and lower vibration-rotation states; and $(d\sigma/d\Omega)_j$ is the cross-section for spontaneous Raman scattering.

On the assumption that the molecules have Boltzmann distributions based on the rotational ($T_r$) and vibrational ($T_v$) temperatures, $\Delta j$ can be expressed as

$$\Delta j = \frac{(2J + 1)g_j}{Q_v Q_r} \left[ \exp\left(\frac{-F_{v,J} \hbar c}{kT_r}\right) - \exp\left(\frac{-F_{v+1,J} \hbar c}{kT_v}\right) \right]$$

(5)

where $F_{v,J}$ and $G_v$ are the rotational and vibrational energy terms, respectively; $g_j$ is the spin degeneracy, 6 for even-$J$ rotational levels and 3 for odd-$J$ levels in $N_2$; and $Q_v$ and $Q_r$ are the rotational and vibrational partition functions, respectively.

From equations (1), (3) and (4), CARS signal is approximately proportional to the square of number density of molecule. Therefore, it is very difficult to detect the CARS signal from low-pressure and strongly radiating gas of hypervelocity.

As written in the following section, we have been successful to detect the CARS signals from hypervelocity air flow. The measured CARS spectra are fitted with calculated CARS signals by treating $T_v$, $T_r$, $G$ as free parameters to decide the temperatures.
3. EXPERIMENTAL APPARATUS AND DIAGNOSTIC SYSTEM

A free-piston, double-diaphragm shock tube has been used to generate strong shock waves in low-density gas as shown in Fig.1. This shock tube consists of high-pressure chamber (driver gas is nitrogen), compression tube (helium gas is supplied) and a free piston, buffer tube (Supplied gas is also helium), low-pressure tube (test gas is air or nitrogen), and vacuum chamber. These tubes are divided off by a quick action valve, the first diaphragm (steel), and the second diaphragm (aluminum). The cross section of the low-pressure tube is 40 mm x 40 mm square. The observation windows of the test section are mounted near a focal lens with some distance from the sidewall of the shock tube. This distance prevents high power laser from destroying the window. The sidewall of shock tube has two small halls along the optical path of laser beam. The shock velocity is measured by ion probes mounted on the sidewall of the test section.

Figure 2 shows a layout of diagnostic system for the CARS measurement in shock waves (CARS system). CARS system consists of a second harmonics Nd: YAG laser ($\omega_1$), a dye laser ($\omega_2$), optical systems, a spectrograph, and an ICCD camera. The laser beam ($\omega_1$) is divided in two by a beam splitter (BS). These beams are directed to the dye laser beam ($\omega_2$) by a beam combiner (BC2). Then three laser beams are crossed and focused with the desirable angles in the shock tube observation section. The CARS spectra are detected by the spectroscopy ICCD camera. The entrance slit width is set to 100 $\mu$m throughout our observation. The ICCD is mounted on the focal exit of the spectrograph.

![Fig. 1 Free-piston, double-diaphragm shock tube]
Fig. 2 Layout of CARS measurement system

Code: HN: He-Ne laser, M: mirror, BC: beam combiner, L: lens, BS: beam splitter, DCM: diachronic mirrors, BP: beam pocket, PH: pinhole, IP: Ion Probe, PT: pressure transducer, DPG: delay pulse generator SG: spectrograph, OS: oscilloscope, LT: low-pressure tube, BT: buffer tube, ST: shock tube.

4. RESULTS AND DISCUSSION

In this experimental paper we indicate the typical four CARS spectrum data of the multiphase flow behind strongly shocked air. The experimental and theoretical spectrum data are shown in Fig. 3~5. The velocity, Mach number and measuring position of about 12mm behind shock front of each shock wave are listed on the Table 1 below.

Table 1 Details of typical measured shock waves

| Fig | Velocity of shock wave [km/s] | Mach number | Measuring position behind shock front [mm] | Fitting error |
|-----|-------------------------------|-------------|-------------------------------------------|--------------|
| 3   | 4.27                          | 12.56       | 12.10                                     | $5.07 \times 10^{-4}$ |
| 4   | 4.27                          | 12.56       | 12.13                                     | $4.69 \times 10^{-3}$ |
| 5   | 4.19                          | 12.32       | 11.90                                     | $1.18 \times 10^{-2}$ |
| 6   | 4.14                          | 12.18       | 11.76                                     | $1.76 \times 10^{-2}$ |

The spectrum data in Figs. 3 and 4 were measured under almost similar conditions. The calculated vibrational and rotational temperatures in Fig. 3 are estimated 7000K, on the other hand, in Fig. 4, the vibrational and rotational temperatures can be estimated 6000K. These results indicate that the N₂ molecules behind shock front are in strong radiation equilibrium but thermally non-equilibrium, and the vibrational and rotational temperatures behind shock front of 4.27km/s are expected between 6000K and 7000K.
On the other hand, the spectrum data in Fig.5 and Fig.6 are different spectrum shapes from Fig.3 or Fig.4. The vibrational and rotational temperatures of Fig.5 are calculated 5500K and 7000K, respectively, while in Fig.6 they are calculated 3000K and 3000K, respectively. The variety of these values are typically detected at the 12mm behind the shock front. It seems that the results in Fig.6 are influenced the cold driver helium gas may be mixed in the buffer tube. Through these results, activated high temperature non-equilibrium gas and low temperature gas which influenced from cold driver gas are mixed in this measuring position. In order to clarify these results around the 12mm behind shock front, we need to obtain the total image of shock waves simultaneously at the velocity about 4km/s.

The fitting error of Fig.4 is the smallest in the four results. Figure 4 seems to be, therefore, more reliable than other data. There is a room for recalculate the spectral data to reduce the fitting error in Fig.5 and Fig.6.

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**Fig.3** CARS spectra (12.10mm behind)  
**Fig.4** CARS spectra (12.13mm behind)  
**Fig.5** CARS spectra (11.90mm behind)  
**Fig.6** CARS spectra (11.76mm behind)
5. CONCLUSION
In this research we have obtained the four cases of spectral data and the vibrational and rotational
temperatures behind the almost fixed conditions of strong shock wave with CARS measurement
system. These spectral results have shown some difference in spite of almost similar experimental
conditions. From these results, the multiphase flow of cold driver gas, radiating plasma, and
vibrationally and rotationally excited gases exists around 12mm behind shock wave of the Mach
number 12. We can determine the temperatures from direct CARS measurement, though, the fitting
figures include a little gaps between experimental spectrum data and theoretical spectrum data. So, we
have to try more precise fitting to clarify the reason why the calculated temperatures are steady and
different from translational temperature.
More precise and larger numbers of experimental data shock waves under the variety of conditions
near to the reentry surroundings need to be obtained.
To support these activities, it’s necessary to take some image photographs of the shock wave and
flow.

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