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Molecular tagging velocimetry of NH fluorescence in a high-enthalpy rarefied gas flow

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Abstract In this paper, a new type of molecular tagging velocimetry based on NH fluorescence was developed and validated for the velocity measurements of a high-enthalpy rarefied gas wind tunnel where the maximum flow velocity exceeds 6 km/s near the nozzle exit at 0.2 Pa. The feasibility of this technique using the short-lived NH fluorescence was demonstrated in the hypersonic rarefied gas flow with yielding velocity profiles at multiple downstream locations from the nozzle exit. The total uncertainty of the measured flow velocities was estimated to be less than 6% when the flow velocity is above 2000 m/s. For the new tagging technique, only a single laser and a single time-gated camera are required for velocity measurement, due to the existence of NH radicals in the arc-discharged N_2 mixed with a small amount of H_2. Therefore, the NH-MTV not only shows great promise for tagging in high-enthalpy rarefied gas of nitrogen or air flows without seeding, but also possesses high practicability and facility for velocity measurement.

Keywords Molecular tagging velocimetry · NH fluorescence · High-enthalpy · Rarefied gas

1 Introduction

In recent years, non-intrusive measurements of highly rarefied gas flows are strongly demanded for the development of vacuum science and aerospace engineering. Especially in hypersonic aerothermodynamics, the enormous progress of numerical analysis using direct simulation Monte Carlo (DSMC) points out the need for accurate non-intrusive velocimetry measurements to validate computational codes and to advance the next generation of hypersonic space transportation systems. Nevertheless, the ability to carry out velocity measurements in a hypersonic gas flow could be rather complicated by the extreme dynamic and thermodynamic conditions present in hypersonic test facilities and the limited optical access to the test section [1]. Furthermore, quantitative measurements are even harder in highly rarefied gas flows where the density is too low to be measured by the ordinary optical interferometry [2]. Consequently, the velocities of hypersonic rarefied gas flows are still hard to measure directly. A variety of non-intrusive methods have been applied successfully to a wide array of flows for measuring velocity [3–10], such as particle image velocimetry (PIV), Laser Doppler velocimetry (LDV), phase Doppler anemometry, and molecular tagged velocimetry (MTV). These techniques fall into three categories: particle-based techniques [3, 4], Doppler shift methods [5–7], and molecular tagged velocimetry [8–10]. In particle-based velocity methods, such as PIV, LDV, and planar Doppler velocimetry, the signals rely on scattering of light from particles present or seeded in the flow. However, the tracer particles do not always follow the flow, especially in hypersonic gas flows, because of large inertia. In Doppler shift methods, it requires the knowledge of line shapes and flow characteristic, sufficient Doppler shift, and strong signal intensity to get velocity. As a consequence, methods in these two categories could not be used in hypersonic rarefied gas flows due to particle lag or insufficient signal strength issues. Thus, molecular tagging velocimetry

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(MTV) becomes a preferable alternative to measure velocity in hypersonic rarefied gas flows.

Molecular tagging velocimetry is a "time-of-flight" technique. The velocity measurement can be easily performed by writing a tag line into the flow and recording its displacements after multiple time delays with no need for the complex calibrations or corrections as in the Doppler shift method. Unfortunately, most of MTV (i.e., OH-MTV, N_2^+-MTV, O_3-MTV) require two or more independently tuned and timed pulsed laser sources. One or two "writing" lasers are used to produce the target molecules by photodissociation or several non-linear techniques such as Raman excitation plus laser-induced electronic fluorescence (RELIEF) [8], two-photon dissociation [9], air photolysis and recombination [10]. The other one is used to excite the target molecules to fluorescing upper states, called the "reading" laser. Meanwhile, some researchers have developed several single-laser MTV methods such as the MTV methods based on acetone [11], biacetyl [12], and nitric oxide [13]. In this case, the "writing" lasers were left out as well as some of the costs of equipment. Meanwhile, the demand of space also can be reduced.

Although a variety of MTV methods based on various tagged species, such as iodine [11, 12], acetone [13], biacetyl [14], nitric oxide [15], have been applied successfully to a wide array of flows such as subsonic/hypersonic flows(<3 km/s) [13–16], microscale flows (~15 μm) [17–19], or the high temperature (~1100 K) gas flows in internal combustion engine [20], very few studies were reported in rarefied gas flows. Moreover, the few studies are concentrated on the low temperature rarefied gas flows (13–200 Pa, at room temperature) by iodine-MTV [11, 12] or the microscale rarefied gas flows by DSMC simulations [13].

As regards the high-enthalpy hypersonic rarefied gas flows (~15–35 MJ/kg), up until now, there are no studies of MTV techniques in the literature on velocity fields, as far as we know. The reason is that most of the traditional tagging species cannot survive in the arcjet (i.e., iodine, acetone, biacetyl, nitrogen dioxide [21]). Some of them encounter particle drag or non-uniform issues (such as sodium [22], strontium [23]) in hypersonic rarefied gas flows. Moreover, many tagging species exhibit strong quenching effects in high-enthalpy flows, giving the insufficient fluorescence lifetime or signal strength [24–26] due to the limits of fluorescence properties.

In this case, we investigate a new type of flow tagging velocimetry technique for high-enthalpy hypersonic rarefied gas flows using the short-lived NH fluorescence in the present experiment. The NH radicals were employed as the target molecules which present naturally in rarefied gas flows all the time by arc discharging in mixed gases of N_2 and small amounts of H_2. Hence, the single "reading" laser is sufficient to tag NH radicals for velocity measurement. Therefore, for the new tagging velocimetry of NH fluorescence, only a single laser and a single time-gated camera are required for the velocity measurement. In situ experimental results indicate that the NH-MTV not only possesses high practicability and facility but also shows great promise for tagging in high-enthalpy nitrogen or air flows without the need for seeding.

2 Experimental setup

The flow tagging measurements were performed in a rarefied gas wind tunnel located at the Institute of Mechanics, Huairou District, Beijing, China. The schematic of the experimental system is illustrated in Fig. 1. The wind tunnel is composed of a gas supply system, a gas heater with its power unit, a planar laser-induced fluorescence system, and a test chamber which is equipped with a cryogenic pump for vacuum evacuation. The continuous rarefied gas wind tunnel is a ground-based simulation facility for the real rarefied gas environment at an altitude of ~100 km. For the purpose of fulfilling the similarity of rarefied state simulation, in experiment, the flow rate of nitrogen (working gas) is set at 0.08 g/s. Hydrogen is added to nitrogen flow with 6% volume fraction for visualization. The mixed gas is heated and ionized by a direct current arc discharge arcjet which is the upgrade one whose structure has been described in detail elsewhere [27, 28]. Estimated power input is about 5 kW. Arc heated gas is accelerated through a laval nozzle with expansion ratio of 400. The nozzle exit is 20 mm in diameter. The test chamber is kept with

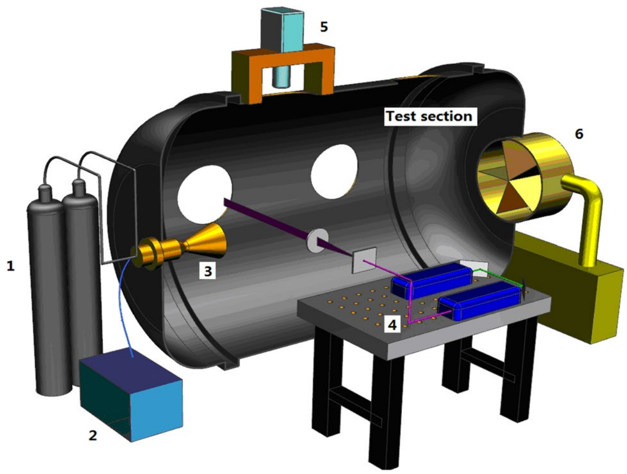

Fig. 1 A schematic diagram of experimental setup and measurement. *1* gas supply system; *2* power unit for gas heater; *3* gas heater; *4* laser and optical arrangement; *5* ICCD camera; and *6* evacuation system

a pressure of 0.2 Pa through continuous pump during the experiment.

It is necessary to note that the continuous rarefied gas wind tunnel is a high-enthalpy hypersonic rarefied gas flow. The simulated altitude is about 100 km away from the ground. The enthalpy value is designed as high as 35 MJ/kg, while the Kunsen number, Mach number, temperature, and the flow velocity in test region are estimated in the range of 0.1–1, 12–20, 300–4000 K, and over 6000 m/s, respectively. Meanwhile, the flow density obtained by analyzing a small ball pendulum in the hypersonic rarefied gas flow varies between 10^{-7} and 10^{-6} kg/m^3 in the tagged region. It means that the density of the tagged radicals may only be in the order of 10^{10} molecules/cm^3. Consequently, the measurement for the high-enthalpy hypersonic rarefied gas flow is a huge challenge, even for the laser-induced fluorescence technique whose detection limit is about 10^{12} molecules/cm^3 according to the reference [1].

For MTV measurement, the NH radicals produced by high-voltage arc discharges were excited by the suitable wavelength ultraviolet laser (around 305.3 nm, at the (1, 0) Q branch-head of the NH $A^3\Pi$-$X^3\Sigma$ transition [29]). The laser was generated from a tunable dye laser source (Model: PSCAN-D-24, Spectra-Physics, dye: Rhodamine B mixed with Rhodamine 610, BBO second harmonic generator) pumped by the second harmonic output of an Nd:YAG laser (Model: Quanta-Ray Pro250, Spectra-Physics). The output laser (the single pulse energy was set around 5 mJ, pulse width is about 10 ns) was vertically expanded to a 150-mm-high laser sheet using a cylindrical lens ($f=-500$ mm) and a long focal length spherical lens ($f=1000$ mm). Before entering the wind tunnel horizontally, the laser sheet was shaped to 10 mm high by a wide open slit.

The NH fluorescence emission of $A^3\Pi$-$X^3\Sigma$ (0, 0) transition around 336 nm [30] was observed and recorded by a fast-gated intensified charge coupled device (ICCD, Model: PI-Max 3, 1024 × 1024, minimum optical gate width: 2 ns, Princeton Instruments) with a f 105-mm UV camera lens (Nikon). The ICCD was secured on the top of the chamber aiming down to the jet exit. With this configuration, the LIF of the tagged region appeared as a thin line on the image plane of the ICCD. To block the plasma emission and laser scattering, the ICCD was spectrally filtered to only accept wavelength at 335 nm with 10 nm FWHM. The intensifier is operated with a variable electronic gate width, usually 100 ns, and a delay varying from 0 to 1000 ns for velocity determination. In general, the fluorescence signals were averaged over 100 shots for good signal noise ratio (S/N). The output of the ICCD was transferred to a computer for analysis. The ICCD synchronization was controlled by the Q-switch synchronous signal of the Nd:YAG laser source whose timing jitter is less than 0.5 ns.

3 Results

Before imaging the tagged region for velocity measurements, a high precision ruler was imaged and the number of pixels per mm in both *x*- and *y*-directions of the camera was determined in order to calibrate the image plane of the ICCD camera for the conversion from pixel number to distance (1 mm ~ 4.8 pixel in the present experiment). The acquired images were corrected for pixel non-uniformity, the effects of dark charge accumulation, and natural arc discharge emissions by a pixel-by-pixel subtraction of an image without excited laser. In the same way, the slope of laser sheet to gas flow was also corrected by subtraction of an image which acquired with zero delay to the laser pulse.

As referred above, the flow tagging technique here is a time-of-flight approach for velocity measurements. In this procedure, the flow was tagged and images were taken at a range of delays. The typical NH PLIF tagging images recorded in the rarefied gas flow are displayed in Fig. 2a. The left image of Fig. 2a presents an image acquired at the arrival of the excitation laser pulse (delay = 0). The image is used as the zero displacement location for the velocity measurements. The right image in Fig. 2a was taken with a delay of 700 ns. It can be easily seen from this image that the flowing gases have displaced the tagged region comparing with the left image (delay = 0, zero displacement position). Meanwhile, the curvature of the tagged region in the right image of Fig. 2a also shows the variations of the velocity of the rarefied gas flows. As a note, all the images in Fig. 2 were collected by 100-ns exposure time with 80% of the maximum intensification of the ICCD, while the number of the shots used for the averaging is 100. The details of the flow conditions have already been described in Sect. 2.

To determine the lifetime of the fluorescence, the NH fluorescence signals in the whole tagged region were integrated and plotted for different delay times. The observed decay process of NH fluorescence as well as the bi-exponential decay fitted curve of the experimental data is displayed in Fig. 2b. The fitting results show that decays of NH fluorescence signals are bounded by a fast and a slow exponential decay. The dual exponential decay fit results in $\tau_1 \sim 100$ ns and $\tau_2 \sim 10{,}896$ ns. The former one was considered as the effective lifetime of NH fluorescence of $A^3\Pi$-$X^3\Sigma$ (0, 0) transition in the present experiment, while the latter one was presumed as the time of diffusion. Note that the lifetime of fluorescence is the time at which the intensity of NH fluorescence has decayed to 1/e or 36.8% of the original value, rather than the whole, during time of the fluorescence. Therefore, although the lifetime of NH fluorescence is just only ~100 ns, the signals can be detected for a long time (until delays up to 1500 ns) as shown in Fig. 2b, while the data could be analyzed in 900 ns.

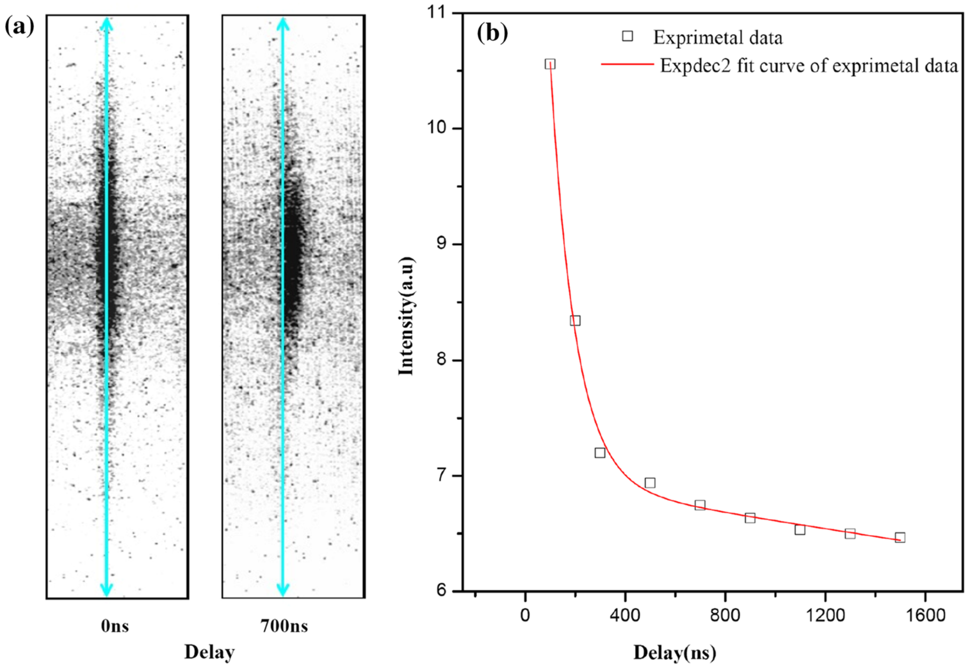

Fig. 2 **a** PLIF tagged images of NH radicals with different delays. **b** The decay process of the NH fluorescence

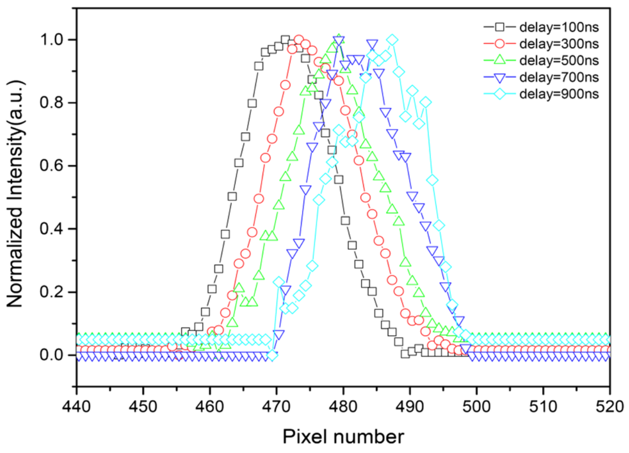

Fig. 3 Variation of the tagged region with time measured in rarefied gas wind tunnel located at streamwise distance is about 45.8 mm from the nozzle

Many previous flow tagging studies [13, 31] have shown that the distribution of the fluorescence at the tagged region could be fitted by least-squares fitting procedure to get quantitative velocity data based on the fitted peak location. Besides, the broadening of the tagged line width (the full width at half maximum of fitted peak) represents the effect of mass diffusion with time. Therefore, the quantitative velocity data could be obtained from the horizontal slices of the images in Fig. 2 that are displayed in Fig. 3. At the same time, Fig. 3 also exhibits the evolution of locations of the tagged region along the gas flow with delay time. The streamwise location is about 45.8 mm from the jet exit ($d=45.8$ mm). In the figure, each curve is a single horizontal slice of the images along the axis of the nozzle, while other experimental conditions are identical to that in Fig. 2. The displacement of the tagged region with the increasing delay times in the flow is clearly depicted in Fig. 3. Peaks of the tagged region were fitted by least-squares fitting procedure with a Voigt spatial profile, similar to the one that has been described in previous flow tagging studies [13, 31]. The proportions of the Voigt spatial profile were optimized by the experimental tagging data. The fitted results shown that the Lorentzian profile account for larger proportion (~70%) in the voigt spatial profile which is also similar to the discovery in previous investigation [13].

In this experiment, the incident laser was narrowed down to 10 mm high before it was introduced into the center region of the flow. If the laser sheet is close to the exit of the nozzle, the tagged radicals would be rather crowed. In this case, the 3D effects which originated from the planar laser sheet and the axisymmetric flow only affect the line width but not the peak position of the tagged line. On the other hand, when the tagged region is far away from the nozzle, the center of the flow is rather uniform because of the high expansion ratio of the laval nozzle (400). Therefore, in this experiment we neglected the 3D effects.

The line width of the fitted curves displayed in Fig. 3 shows that the diffusion of the tagged region which usually appears in flow tagging experiments is not obvious in the present experiment. We attribute this to the high-speed of the gas flows and the short delay time of acquisition. This conclusion can also be confirmed by the time of diffusion

(~10 μs) deduced from the decay of NH fluorescence signals in Fig. 2b.

Then, the centers of the tagged regions can also be obtained by the least-squares fitting for the peaks in Fig. 3. The locations of the tagged region could be determined by the fitted peak values as well as the displacements of the tagged region. The velocities of gas flows are calculated based on the displacements of the tagged region with different delay times. According to the curves in Fig. 3, the displacements of the tagged region in each delay are determined and demonstrated in Fig. 4 with the delay times. Thus, the velocity in this location ($d=45.8$ mm) was obtained by the linear fitting of these displacements with delay times, which gives the velocity of 3826 ± 160 m/s. The strong linear relationship reveals not only the reliability and the accuracy of the flow tagging method based on NH fluorescence, but also the stability and the long-time running characters of the rarefied gas wind tunnel.

Similarly, the velocity profiles of the rarefied gas flows at four different streamwise locations from the jet exit in the direction perpendicular to the flow are calculated and exhibited in Fig. 5. As shown in the figure, each curve represents the velocity profile of corresponding distance from the nozzle. The radial distance denotes the horizontal distance from the central axis of nozzle. The enthalpy value of the Arcjet was increased from ~15 MJ/kg to about 35 MJ/kg for speeding up the gas flow. Therefore, the velocities exhibited in Fig. 5 are significantly higher than the previously displayed data. At the same time, some acquisition conditions were also changed except for the exposure time and the number of shots for averaging. The intensification was set at 90% of the maximum intensification of the ICCD, while the single pulse energy of the incident laser was decreased to 3 mJ/pulse. Besides, each symbol in Fig. 5 is the average result of about 25 rows in the direction along the laser beam to get high S/N ratio.

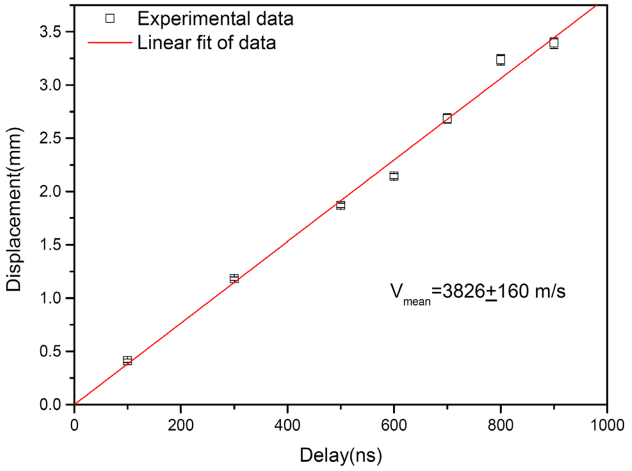

Fig. 4 Measurement of average flow velocity by fitting a straight line through time-of-flight data along axis of the nozzle

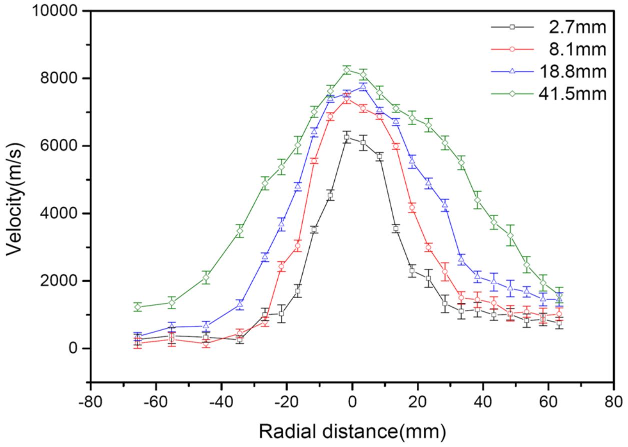

Fig. 5 The distributions of radial velocities for the rarefied gas flow at different locations (gate:100ns, delay:350ns, figure legends are distances of tagged lines from the exit of nozzle)

As displayed in Fig. 5, the measured velocity profiles of the gas flows show that the velocities of the high-enthalpy rarefied gas flow are rather high which exceed 6000 m/s near the nozzle. Then, the velocities increase along the flow direction, while the high-speed gas spread radially to the direction of gas flow. It manifests that the gases jetted from the nozzle are going through an expansion process which coincides with the flow rule of free jets. Besides, the details of the flow field also could be observed that the gas flow is near symmetry in a direction perpendicular to that of stream in the high-enthalpy rarefied gas flow facility. The slight dissymmetry is attributed to the skewing of the electric arc relative to the axial direction of the wind tunnel. The error values of velocity measurement in Fig. 5 are derived from the accuracies of the displacements of tagged region which are the rms sum of the uncertainty of the magnification of the optical system and the statistical uncertainties of the peak locations determined by the least-squares fitting procedure for the sequential tagged images. According to the experiment, the error is strongly dependent on the signal-to-noise ratios and the flow velocity. In this experiment, the signal-to-noise ratios (S/N) of the collected experimental images are usually greater than one even though the delay time of acquisition goes up to 900 ns. In this instance, the statistical uncertainty in this work is typically found to be of the order 4–5% of the displacements of tagged region when the flow velocity is above 2000 m/s. However, the systematic errors in this experiment should include not only the referred statistical uncertainties of the tagged line locations, but also the measurement uncertainties of the magnification of the optical system (<1% in this work) and the timing uncertainties between the laser and camera.

According to the view of Danehy et al. [16], we estimated the total timing uncertainties in our measurement to be less than 1 ns because the time jitter of the laser is less than 0.5 ns, and the timing uncertainty of ICCD (PI-Max3) is only 35 ps. Besides, the effects of radiative heating of the flow and ablating on surface of facility by the laser were negligible, due to the low energy density of the excitation laser and the large diameter of the wind tunnel. Therefore, the total measurement uncertainty of the measured flow velocities in this work is then estimated to be less than 6% when the flow velocity is above 2000 m/s.

4 Conclusions

We have investigated into the application of a new flow tagging technique based upon NH fluorescence for velocity measurements in a highly rarefied gas wind tunnel which provides high-enthalpy flows with maximum velocity greater than 6 km/s near the nozzle exit. The NH radicals were produced as the target molecules by arc discharging in mixed gases of N_2 and small amounts of H_2. Even though the effective lifetime of its fluorescence in the present experiment is rather short (about 100 ns in the present experiment), the feasibility of this method was demonstrated by the measured flow velocities and its profiles in the direction perpendicular to the flow at different downstream locations from the nozzle exit for the high-enthalpy rarefied gas wind tunnel. At the same time, the need for only one laser and a single time-gated camera also shows very high practicability and facility of the new NH-MTV for velocity measurement.

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