Laser Ion Time-of-Flight Velocity Measurements Using N_2^+ Tracers

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A laser ion time-of-flight technique for velocity measurements using N_2^+ tracers has been demonstrated. The ions are formed by a multiphoton interaction with the third harmonic of a Nd:YAG laser and are detected by a negatively biased ion probe. In air flows with positive probe bias, negative ions, possibly O_2^- formed by attachment with free electrons, were detected and could also be used as tracers. The lifetime of the ions is controlled by charge recombination. At atmospheric conditions and after a delay of 600 μ s, ion signal was still above background noise level. Measurements of flow velocities in both a low-speed freejet and in the flame of a propane-air torch are presented. Reduced flight time in high-speed flows can produce even higher signals.

Introduction

■ HE validation of numerical codes that simulate high-speed . mixing and combusting flows is considerably enhanced by velocity measurements. Velocity measurements are desirable at the entrance plane, in the mixing and combusting region, and at the exit of supersonic combustors. Velocity measurements are also required in subsonic and supersonic air flows. Although a number of techniques, with varying degrees of complexity and success, have been applied for these measurements, none can be applied, in reacting and nonreacting flows, without seeding of the flow. A technique, using multiphoton ionization of N₂ followed by time-of-flight (TOF) detection using an electrically biased ion probe has been demonstrated successfully in air and nitrogen flows and in the flame of a propaneair torch. This laser ion time-of-flight (LITOF) technique using N₂, a specie with global presence in aerodynamic flows, does not require seeding of the flow, can be used over a wide range of density, and is applicable to reacting and nonreacting air flows. The technique is relatively simple to implement and should be applicable in environments having moderate velocity gradients and where probe survivability and intrusion is not a constraint. In addition, in tests involving aerodynamic models, the ion probe can be incorporated in the model surface, thereby rendering the technique nonintrusive.

Techniques for velocity measurement in high-speed flows can be listed under two general categories: 1) measurements sensitive to the Doppler shift experienced by radiation interacting with high-speed molecular species or particles and 2) time-of-flight techniques that depend on the displacement of a tracer over a prescribed path length and time interval. Among the most well-developed approaches is laser Doppler velocimetry (LDV).1 This technique uses the measured Doppler shift of radiation scattered from particles in the flow to establish velocity. A more recent particle scattering technique, particle imaging velocimetry (PIV),² uses a time-of-flight approach to determine velocity from particle displacement obtained from successive images. Both LDV and PIV techniques require particle seeding and, hence, may suffer from systematic errors when the particles fail to track large velocity gradients. This is particularly serious for velocity measurements in high-temperature flows where dense refractory material particles must be used. In addition, seeding can influence reaction kinetics in reacting flows. Alternatively, the Doppler shift associated with the absorption and scattering process in molecular species can be used for velocity measurements. Species previously used for these measurements include Na (Ref. 3), I₂ (Ref. 4), NO (Ref. 5), and OH (Ref. 6). However, these species are not naturally present in nonreacting air flows. Although OH and NO may occur naturally, their presence is usually limited to areas downstream of the reaction zone of combusting flows. Furthermore, Na, I₂, and NO may interfere with the reaction if artificially seeded to obtain velocity in combusting flows.

Alternative velocimetry techniques using the TOF approach include the formation of a thermal lens⁷ by locally heating the flow with a laser pulse, excitation of a long lived molecular specie such as the first vibrational level of O₂ (Ref. 8), or the dissociation of a molecular specie, e.g., H₂O (Ref. 9) by an uv laser pulse. In these measurements the location of a tracer formed by the laser pulse is determined after a delay by an optical imaging technique using a second laser beam. The velocity is determined from the measured distance between the point of formation of the tracer and its location at the time of measurement and from the measured time between the two events. Unfortunately, the thermal lens technique may not be applicable to flows where large temperature and density gradients exist. The application of an oxygen-based TOF technique is generally limited to the nonreacting and low-temperature regions of air flows, whereas the application of the H₂O dissociation technique is limited primarily to the reacting regions or requires H₂O seeding. By contrast, this technique offers the potential for velocity measurements both in the reacting and nonreacting regions of the flow without the need for seeding using a relatively simple measurement scheme.

Ion time-of-flight velocity measurements have previously been demonstrated in low-density hypersonic air flows using electron beam ionization of N_2 and an ion probe for detection of the ions. ¹⁰ However, the use of an electron beam for the formation of N_2^+ limits the technique to low-density air flows. Velocity has also been measured in UF₆ using laser induced gas breakdown for ionization in a freejet¹¹ and for determining azimuthal, axial, and radial velocity components in a full-scale gas centrifuge. ¹² Thus, although the use of an ion probe for velocity measurements using a TOF approach has already been demonstrated, the present work is the first demonstration of a nitrogen LITOF technique. Use of a laser pulse for the formation of the ions removes the limitations associated with the use of an electron beam thereby becoming useful for a wide range of velocity measurement needs.

Theoretical Background

The multiphoton laser-induced N_2 ionization process was demonstrated recently.¹³ In this experiment the excitation of N_2 in atmospheric air by the third harmonic of a Nd:YAG laser was observed to result in an emission at 391.4 nm. This emission could not be observed when irradiating pure O_2 with the same laser. The emission

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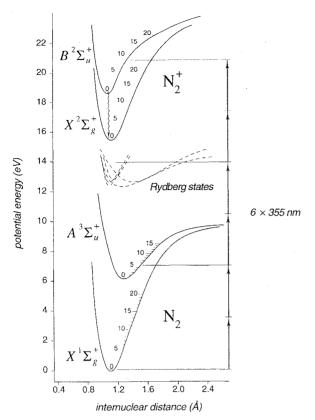


Fig. 1 Energy level diagram for the six photon ionization of N_2 , adapted from Ref. 14.

spectrum was analyzed spectroscopically and was determined to result from transitions in the $B^2\Sigma_u^+ \to X^2\Sigma_g^+$ band of N_2^+ . Vibronic transitions originating from v'=0,1,2,3,4, and 5 were identified, with the most intense emission at 391.4 nm resulting from the $v' = 0 \rightarrow v'' = 0$ transition. Energetically, six photons at 355 nm are required for the excitation of the $B^2 \Sigma_{\mu}^+(v'=8)$ level. But, since the intensity of the 391.4-nm line was found to increase quadratically with the incident pulse energy, a multiphoton ionization process involving three intermediate two-photon steps (see Fig. 1) was hypothesized. 13 The total number of ions formed by a 45-mJ laser pulse was estimated 13 at $\sim 10^7$, corresponding to a density of $\sim 10^{13}$ /cm³. Thus, although the energy absorbed by each ionized molecule exceeds 20 eV, the average energy deposited by the laser beam in atmospheric air is less than 2×10^{-5} eV/molecule resulting in a temperature rise of $\ll 1$ K even if the entire ionization energy were thermalized. Although the lifetime in the $B^2\Sigma_{\mu}^+$ electronic level is short relative to the laser pulse duration, the lifetime at the $X^2 \Sigma_a^+(v''=0)$ level is indefinite and is determined solely by the charge recombination rate or by the charge transfer rate between N_2^+ and other species of the flow such as O_2 .

In pure nitrogen, the primary mechanism for the removal of ions is expected to be dissociative recombination through the following reaction:

$$N_2^+ + e^- \to N + N \tag{1}$$

Assuming that at any time the density of the ions is identical to the density of the free electrons, the variation with time of the ion density can be found by solving the rate equation. Thus, for an initial density $[N_2^+](0)$, the density at time t is

$$[N_2^+](t) = \frac{[N_2^+](0)}{[N_2^+](0)k_Dt + 1}$$
 (2)

where $k_D \approx 1.06 \times 10^{-6}$ cm³/s (Ref. 15) is the charge recombination rate.

In air, the primary removal mechanism of N_2^+ is by charge transfer with O_2 through the following reaction 16 :

$$N_2^+ + O_2 \rightarrow N_2 + O_2^+$$
 (3)

With a rate of 6.0×10^{-11} cm³/s, at atmospheric conditions, most of the charge is exchanged within a time that is short relative to the duration of the laser pulse. Other components of atmospheric air such as $\rm H_2O$ and $\rm CO_2$ may also participate in charge transfer reactions with $\rm N_2^+$. However, a further charge transfer between $\rm CO_2^+$ and $\rm O_2$ at a rate of $\rm 10^{-10}$ cm³/s (Ref. 17) and between $\rm H_2O^+$ and $\rm O_2^+$ at a rate of $\rm 2 \times 10^{-10}$ cm³/s (Ref. 18) assures that the positive charge transfer that is predicted by Eq. (3) is not inhibited. There is no charge transfer between $\rm N_2^+$ and argon, $\rm ^{17}$ also a major constituent of atmospheric air. Although TOF measurements using an ion probe, where only the charge and not its molecular identity is determined, are not affected directly by this transfer mechanism, the identity of the ion may influence the choice of spectroscopic options that may be used for future measurements.

Similarly to the positive charges, the fate of the negative charges depends on the gas constituents. In pure nitrogen the free electrons are removed primarily by dissociative recombination with N₂ [Eq. (1)]. In atmospheric air, free electrons are likely to attach to O₂ which has an affinity of 0.46 eV for an attachment reaction. 19 Typically, the attachment is controlled by a three-body collision where the third body is needed for deactivation. For deactivation by O2, N_2 , and CO_2 the rates²⁰ are 4.1×10^{-30} cm⁶/s, 1×10^{-31} cm⁶/s, and 2.8×10^{-30} cm⁶/s. Although higher order collisions were also identified as contributing to the attachment reaction.²⁰ the rates for three-body recombination are by themselves sufficient to attach, at atmospheric conditions, the entire population of free electrons to O₂ within a time that is short relative to the duration of the laser pulse. Thus, instead of a recombination with the relatively light free electrons, charge recombination in air is controlled by molecular collisions where all of the charges, positive and negative, are attached to O₂. Consequently, the charge recombination rate in air is expected to be slower than the recombination in pure nitrogen. Ultimately, the lifetime of the ions both in N₂ and air is determined by the rate of charge recombination. Therefore, relatively long lived ions can serve as tracers for TOF velocity measurement. With this technique, ions are formed by the ionizing laser beam at a prescribed location and are subsequently detected either spectroscopically or by an ion probe at another location. The distance traveled by the ions and the measured time are used to calculate the velocity.

Apparatus and Procedure

This nitrogen LITOF technique has been demonstrated using the third harmonic of a Nd:YAG laser to form the ions and an electrically biased probe for their detection. Velocity measurements were obtained in air or nitrogen subsonic flows and also in the flame of a commercially available propane fueled torch. Detection of the ions with a probe allowed careful measurements of characteristics such as the variation of ion density with the laser pulse energy, confirmation of the charge recombination mechanism that controls lifetime, the distances that can be traveled by the ions with the flow without dissipation, or the velocity measurement limitations.

Figure 2 is a schematic of the experimental setup used for these measurements. The third harmonic of a Q-switched Nd:YAG laser,

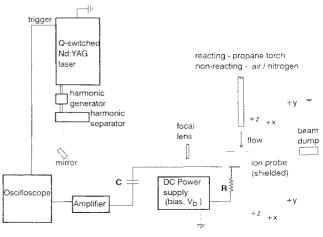


Fig. 2 Experimental setup.

operating at 10 Hz, was focused into a nominal diameter of 17 μ m. To avoid gas breakdown, the third harmonic component was separated from the first and second harmonics and limited to an energy of 30 mJ.

For measurements in cold flows, an ion probe consisting of a stainless-steel shielded needle with just the metal tip exposed was placed downstream of the focus of the laser beam on a three-axis translation stage. The needle was aligned parallel to the flow, thereby providing an excellent spatial resolution nonetheless minimizing perturbation of the flow. Furthermore, owing to its pointed shape, the high-electrostatic field at the tip of the probe exceeded the field at the tagged volume even after the removal of most of the laserinduced free charges that survived recombination at the time of the measurement (<10⁶ charges). Also, owing to the capacitance of the electronic system, attachment of $<10^{6}$ charges perturbed the probe voltage by <0.001 V. For measurements in reacting flows, the probe consisted of a platinum wire passing through an electrically shielded ceramic tube with a section of 1 mm exposed. This probe was placed normal to the main flow direction to assure that the flow does not penetrate the gap between the probe and the ceramic shield. In both measurements, the laser beam was passed normal to the flow and upstream from the ion probe.

The nonreacting jet consisted of either air or nitrogen (99.998% purity) flows exiting from a straight section of a stainless-steel tubing with an exit diameter of 8.4 mm and a length to diameter ratio, l/D=45. With this ratio the velocity profiles appeared to be fully developed. The total flow rate was determined independently by measurement of pressure drop across a choked orifice. In addition to its simplicity, this subsonic jet allowed velocity measurements in situations that require relatively long TOF thereby affording an accurate characterization of most of the ion properties. Furthermore, since the lifetime of the ions is expected to be the primary limiting parameter for this technique, measurements in subsonic flows are likely to establish the velocity measurement limitations.

Measurements in reacting flows were demonstrated in the core section of a commercially available propane air torch. For calibration of the velocity measurements, a fuel flow rate of 1.04 g/min was measured gravimetrically.

The detection circuitry consisted of a 360-pF capacitor, connected in series, to the probe. This capacitor was used to transmit the transient currents while isolating the electronic detection system from the dc bias voltage. The current past the capacitor was passed through a 110-K Ω load resistor. The voltage generated across this resistor by the transient current was amplified and displayed on the screen of a high-impedance storage oscilloscope triggered by the Q-switch controller of the laser. Because of the pulsed nature of this experiment and the relatively low number of charges available for measurement (<10⁶), the system response was characterized by the time constant which for this setup was relatively long (\sim 5 μ s). However, it was short relative to the characteristic time of this experiment which exceeded 100 μ s.

Results and Discussion

Figure 3 presents a typical oscillogram of the time-dependent variation of the voltage across the load resistor for air flow when the probe was negatively biased and placed at a distance of 3.8 mm downstream from the laser focus. This is similar to previously reported¹⁰ oscillograms obtained by the ionization of a hypersonic air flow by an electron beam. In Fig. 3 two pulses can be noted. The first pulse, which was detected almost simultaneously with the incident laser pulse, decayed at a rate which was consistent with the time constant of the system. The second pulse was delayed relative to the first pulse with the delay time increasing linearly with the separation between the probe and the laser beam and also with the velocity of the flow. Therefore, the second pulse could be attributed to the laser-induced ions transported by the gas flow to the probe. A flow velocity of approximately 50 m/s was determined from measurements of this delay and of the separation between the probe and the focus of the laser beam.

To determine the source of the initial pulse in Fig. 3, the variation of its integrated area with the incident laser energy was measured. A quadratic increase of the area of this pulse with laser energy

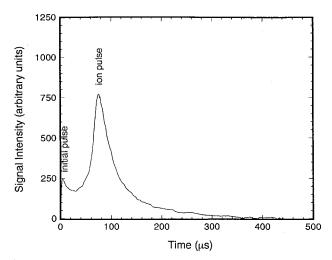


Fig. 3 Typical oscillogram of the time-dependent variation of the voltage across the load resistor for air when the probe is negatively biased.

was observed. This is similar to the previously reported 13 quadratic dependance between the density of the laser-induced N₂⁺ ions and the incident laser energy. This result suggests that the magnitude of this pulse is proportional to the initial density of the ions. In addition, since it was recorded almost instantaneously it was associated with electrostatic effects induced by the electric field of the probe. This conclusion is supported by the absence of this initial pulse when the probe is fully shielded¹¹ or when the separation between the probe and the ion source is large. 12 Although in the present experiment the initial pulse was consistently detected it was found to decrease dramatically with separation between the probe and the focus of the laser beam. At a distance of 1 mm between the needle probe and the laser beam, the magnitude of this initial pulse was only 5% of the magnitude at a separation of 0.3 mm. Thus, at separations in excess of 1 mm the effect of this pulse on ion TOF measurements is expected to be negligible. This initial pulse is also consistent with the electron detection pulse that was observed in a previous resonant-enhanced multiphoton ionization (REMPI) experiment²¹ where a probe was used for the detection and measurement of the density of NO. By translating the ion probe parallel to the laser beam and monitoring the initial-pulse amplitude, the length of the volume tagged by the laser-induced ionization was estimated to be

The previous experiment was repeated both in air and nitrogen flows with the ion probe positively biased. In air flows the oscillograms were similar to that in Fig. 3 with both the initial and the delayed pulses present. The presence of the delayed pulse in air flows when the probe is positively biased suggests that along with the formation of N_2^+ , a negative ion, possibly O_2^- , is also formed. This is consistent with the reported²⁰ high-attachment rate of lowenergy electrons to O_2 which would result in the formation of O_2^- at a density comparable to the initial density of N_2^+ within a time that is comparable to the laser pulse duration. This hypothesis was confirmed by conducting similar experiments in a pure N_2 flow. When the probe was negatively biased, the oscillogram was similar to those obtained in air. However, when the probe was positively biased, only the initial peak due to the electrostatic effects occurred, with no evidence of a transported ion peak.

Before this technique could be applied for velocity measurements, the effect of the acceleration imparted on the ions by the electric field of the probe had to be determined. Since the acceleration induced by the probe depends on the probe geometry and the probe potential, measurements were required both in the reacting and nonreacting flows where different probe geometries were used. Figure 4 presents the variation with probe voltage of the time of arrival of the ion pulse in the propane torch flame, while maintaining the fuel flow rate and the separation between the laser beam and the probe at 3.2 mm. Clearly when the bias voltage exceeds 10 V, the probe potential can significantly affect the ion flight time. Therefore, velocity measurements using this probe were performed with a bias voltage of 5 V where potential induced acceleration is negligible. The effect

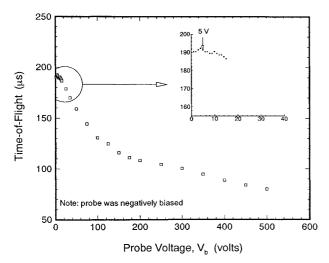


Fig. 4 Variation of the time of arrival of laser-induced ions in a propane air flame with the probe bias voltage.

of the probe potential on the ion TOF in nonreacting flows, where a needle tip probe oriented parallel to the flow was used, was less pronounced. With a separation of 2.5 mm between the probe and the laser beam, the effect of a probe voltage of 150 V on the ion TOF was measured to be less than 5% for the velocity of the current study. The relatively small effect of probe voltage on the ion velocity, particularly when the pointed needle probe was used, is an evidence that the electrostatic field decays rapidly away from the probe. Therefore, the collection cross section of the probe is determined primarily by its physical dimension. If a similar acceleration of 5% is imparted on the transverse velocity component, it may be assumed that with the exception of that error ions not intercepted directly by the probe are not collected. To measure the effective dimensions of the collection cross section the probe was traversed longitudinally and transversely to the laser beam while monitoring the ion signal. Transversely the probe could be moved approximately ± 1 mm from the beam centerline before the signal was lost. Although the combined size of the probe and the beam are <1 mm, turbulence may distort the shape of the ion line²² thereby effectively increasing the collection cross section. Longitudinally to the beam, the length of the probe volume was approximately 6 mm, thereby suggesting that to be the length of the ionized volume.

Since the measurement of the velocity requires independent measurements of the ion TOF and of the separation between the laser beam and the probe, the resolution is ultimately limited by the uncertainty in these measurements. In most applications, the error associated with the measurement of the TOF is expected to be dominant, with the resolution of the velocity measurement limited by the width of the transported ion pulse. To estimate the potential of this technique for measurements in high-speed flows the variation of the full width at half the maximum (FWHM) of the transported ion pulse with flow velocity was determined. Figure 5 presents the variation of the FWHM with ion TOF in air flow obtained by maintaining the separation between the laser beam and the ion probe constant although changing the flow velocity. It is seen that at low velocities, i.e., at relatively long TOF, the FWHM of the ion pulse increases linearly with the TOF. Such behavior is possible when the width is controlled by the time required by the volume containing the ions to sweep the exposed section of the probe. Thus, as the velocity increases this time decreases thereby reducing the width of the ion pulse. At higher velocities the FWHM appears to scale with (TOF)^{0.4}, approaching the detection circuit limiting time constant at $t \approx 10 \,\mu s$. Thus, the measurement resolution is expected to be limited at high-flow speeds by the time constant of the system. For short flight times that are associated with high-flow speeds, ion signals are significantly increased [Eq. (2)], permitting reduction of detection circuit time constant and, consequently, the widths of both the initial peak and ion signal.

At low speed the velocity measurement using this technique is limited primarily by the lifetime of the laser-induced ions. As the

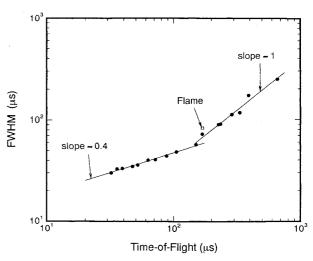


Fig. 5 Variation with the TOF of the full width at half the maximum of the ion pulse when the distance between the probe and the laser focus is constant.

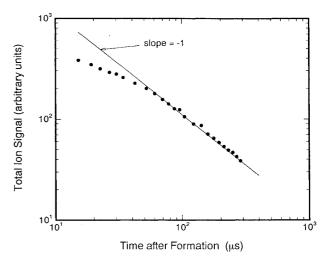


Fig. 6 Variation of the total ion signal with TOF in pure nitrogen flows.

delay between the formation of the ions and detection increases, the number of the ions available for detection decreases [Eq. (2)] until the induced signal falls below the background noise level. Thus, the ion lifetime and the mechanism controlling it must be well defined. To confirm the mechanism that controls the lifetime of the laser-induced ions, the variation with TOF of the total ion charge detected by the ion probe was measured. Figure 6 presents a logarithmic plot of the detected ion signal in pure N_2 vs the flight time. It is seen that when $t>35~\mu s$ the variation of the detected ion density follows a line with a slope of -1, which is predicted from Eq. (2) when $\{N_2^+\}\{0\}k_Dt\gg 1$ whereas at shorter TOF the slope is more gradual, also in agreement with Eq. (2). Similar behavior was also observed for the variation of the detected ion signal in air vs the flight time. The present results also suggested that ion signals are sufficient for detection with an ion probe for flight times $<600~\mu s$.

To demonstrate the potential of this technique, velocity profiles were measured both in reacting and nonreacting flows. Figure 7 presents a series of velocity profiles obtained for a subsonic, turbulent, freejet of air. The profiles were obtained at several locations, x = 1, 4, 6, and 8 tube diameters downstream from the exit plane. At the shear layer, where the velocity was 4.5 m/s or less, the recorded signal intensity of the ion pulse fell below the background noise level. Therefore, the dashed line on each of the velocity profiles indicates the threshold for these velocity measurements. The velocity measurement uncertainty due to uncertainties in the TOF measurements is presented by the error bars. For comparison, an independent measurement of the volumetric flow rate through a choked orifice was obtained. This measurement compared within 10% with the

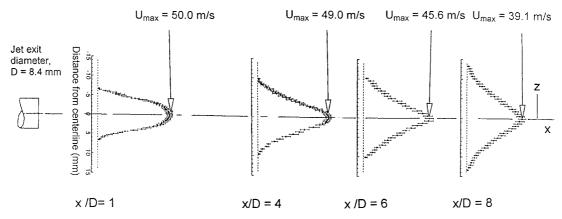


Fig. 7 Velocity profiles obtained in subsonic turbulent freejet of air using the ion TOF techniques at x/D = 1, 4, 6, and 8.

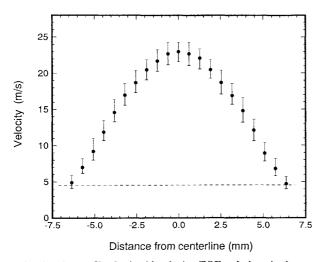


Fig. 8 Velocity profile obtained by the ion TOF technique in the reaction zone of a propane air torch.

numerically integrated velocity profile measured downstream from the jet exit at x = 1D. The slight discrepancy can partially be attributed to the entrainment of ambient air at the shear layer.

To evaluate this nitrogen LITOF technique in a reacting flow, it was applied to a commercial propane/air torch. Figure 8 shows a measured velocity profile obtained 24 mm downstream of the circular 13-mm-diam exit of the torch. No independent measurement of the velocity was available. However assuming that the mixture of fuel and air is stoichiometric and at a total temperature of 1500 K, a fuel flow rate of 1.36 g/min was predicted from the integrated velocity profile. This compares well with the fuel flow rate of 1.04 g/min that was measured gravimetrically.

Conclusions

A TOF technique for velocity measurements in nonreacting and reacting air flows using laser-induced N₂⁺ tracers has been demonstrated. An electrically biased ion probe was used for detection of the ions. Measurement uncertainties due to electrostatic effects induced by the probe field were determined to be small when the separation between the probe and the laser beam exceeded 1 mm. Also, depending on the probe geometry, acceleration of the ion by the probe electrostatic field can be minimized by selection of sufficiently lowbias voltage. Bias voltages of 5 V and 150 V were selected for the probes in this experiment. In air flows, both positive ions, possibly O_2^+ , and negative ions, possibly O_2^- , could be detected. On the other hand, in pure nitrogen flows only positive ions were available for the measurement. The measurement resolution was shown to be limited by the FWHM of the detected ion pulse. However, as the velocity increases the width of the pulse decreases until it is limited by the time constant of the detection system. At low-flow speeds the measurement is limited by the lifetime of the ions. At a delay of 600 μ s or less the signal level was above the background noise level thereby allowing effective velocity measurement. Velocity measurements were successfully demonstrated in a turbulent subsonic freejet of air and in the reaction zone of a propane-air torch. Velocities of greater than 4.5 m/s were resolved and compared favorably with independent flow rate measurements. Nonintrusive ion interrogating techniques, such as laser-induced fluorescence, may be considered as an alterative for the ion probe detection.

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