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Air Photolysis and Recombination Tracking: A New Molecular Tagging Velocimetry Scheme

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A new scheme for molecular tagging velocimetry in unseeded airflows is presented. The method, called air photolysis and recombination tracking, is based on the photoinduced formation of nitric oxide (NO) in the waist region of a focused ArF excimer laser beam. The distribution of the formed NO molecules is imaged by planar laser-induced fluorescence in the γ band, using a frequency-doubled dye laser beam. The role of N_2^+ ions in the NO formation process is discussed, and the lifetime of the NO molecules was determined to be at least 10 ms. The new method has been applied to a laminar and a pulsed airflow and a premixed methane/air flame. Velocities could be determined with an accuracy of 5% in the airflows (on a single-shot basis) and 13% in the flame. By the use of averaging over many laser pulses, velocities as low as 1 cm/s could be measured.

Introduction

IN molecular tagging velocimetry (MTV),[1–3] a pulsed laser is used to write a pattern in a flowfield. After a time delay the pattern, which has been altered by the flowfield, is read back. From the displacement and the deformation of the pattern, a map of velocity vectors can be determined. The information obtained with MTV is comparable to that obtained with particle-based techniques such as particle imaging velocimetry (PIV). However, problems related to the use of tracer particles, such as particle lag, disturbance of the flowfield, etc., do not occur with MTV.

Several techniques that can be used for molecular flow tagging in gas flows have recently been developed. Techniques based on the photodissociation of NO_2 (Ref. 1) or tert–butyl nitrite (Ref. 2) to form nitric oxide (NO) or the photoexcitation of biacetyl (Ref. 3) demand homogeneous molecular seeding of the flow. The written pattern is read by means of laser-induced fluorescence (LIF) of NO or laser-induced phosphorescence of biacetyl, respectively. The application of these methods is limited to closed flow systems, because the additives are toxic or at least irritant. (Biacetyl has a very irritant smell.) Furthermore, biacetyl phosphorescence is strongly quenched by oxygen, and therefore, this method can be used only in the absence of oxygen.

For the investigation of turbulent freejets, a method that does not demand the addition of seed molecules or particles would be preferable. For this purpose, we need a method with the following features: Complicated patterns can be written with a single write pulse and the read process can be performed with a single read pulse with delays between both pulses in the order of relevant timescales of the flow. Therefore, it is very important to find a technique with large signal intensities and a long lifetime of the tagged molecules. Known techniques suitable for unseeded airflows are Raman-excitation and laser-induced electronic fluorescence (RELIEF)[4] of O_2, H_2O photolysis followed by detection of OH (Ref. 5), and photodissociation of O_2 followed by detection of O_3 (Ref. 6).

We have introduced a new method for MTV in unseeded airflows that is based on the photosynthesis of NO (Ref. 7): air photolysis and recombination tracking (APART). In this paper new information on the mechanism of the NO formation is presented, and the lifetime of the tagged molecules is determined. Applications of APART to a laminar and a pulsed airflow are shown. Finally, the applicability of APART in reactive flows is investigated.

Experiments

APART is based on the photosynthesis of NO. The beam of a broadband ArF excimer laser (Λ-Physik, CompeX 350T, used in amplifier-only mode; $\lambda = 193$ nm, pulse length 20 ns, and repetition rate 10 Hz) with a pulse energy of about 40 mJ/pulse was focused in air by an $f = 100$ mm spherical lens, leading to the formation of NO in the waist region by recombining the atoms in the main air constituents O_2 and N_2. The waist of the focused excimer beam was ~0.18 mm, resulting in a laser irradiance of about 16 W/mm^2. A detailed description of APART has been published elsewhere.[7] Optical breakdown should be avoided because it spoils the nonintrusive nature of the technique and no detectable amounts of NO are found after breakdown. A Nd:YAG (Continuum PowerLite 9010; $\lambda = 355$ nm and pulse length 5 ns) pumped dye laser (Λ-Physik, ScanMate 3, operated on Coumarin 2) was frequency doubled to obtain radiation with a wavelength of 226 nm. The 226-nm beam was used to excite the $A(v' = 0) \leftarrow X(v'' = 0)$ system of NO. LIF emission from the A state was detected with an intensified charge-coupled device (CCD) camera.

In the tagging experiments, the dye laser beam was aligned anticollinearly to the excimer beam, perpendicular to the flow direction, and focused into a sheet (in the plane spanned by both laser beams and the flow direction) with a thickness of about 0.2 mm and a height of 10 mm by a cylindrical lens ($f = 300$ mm). In Fig. 1, a schematic of the tagging setup is shown. At $t = 0$, NO molecules are formed in the waist region of the pulsed ArF excimer laser beam. After delay time Δt, the NO distribution is imaged by planar laser-induced fluorescence with a dye laser sheet. The wavelength of the dye laser was fixed to the $Q_1(14) - Q_2(21)$ coincidence at 225.95 nm (indicated with an asterisk in Fig. 2) in the $A(v' = 0) \leftarrow X(v'' = 0)$ system of NO. LIF emission from the A state was detected. A 226-nm, 0-deg mirror (Laser Optik) was used to suppress light with the laser wavelength. The flow area was imaged onto an intensified CCD camera (Princeton Instruments, ICCD-576G/RB-E) through a Nikon f/4.5 105-mm UV objective. The resolution of the imaging system with 4×4 binning on the CCD chip was 38×38 μm^2 per pixel. This corresponds to a field of view of 5.5×3.6 mm^2.

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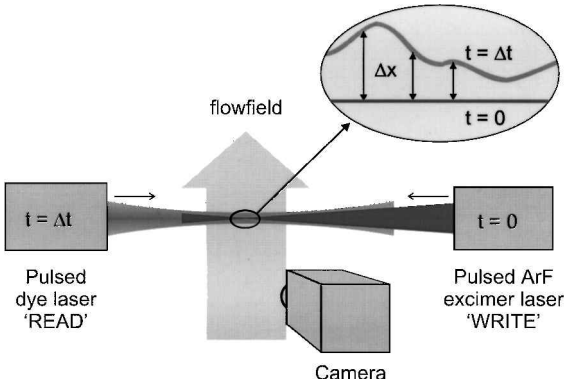

Fig. 1 Setup for APART.

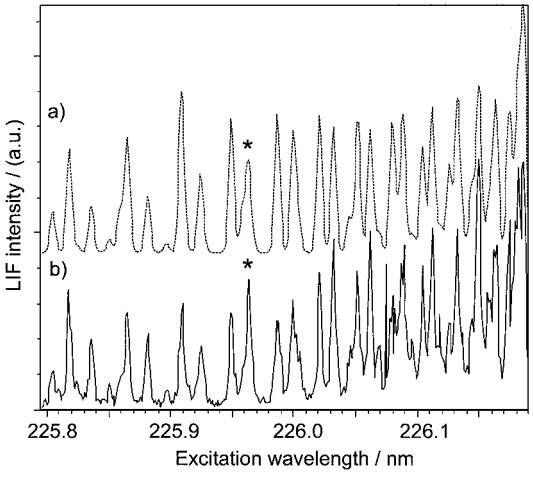

Fig. 2 Spectrum a) of NO (0,0) γ-band excitation calculated with LIFBASE and b) measured in the waist region of the excimer beam (delay between dye laser and excimer laser pulse 1 μs).

Results and Discussion

APART Method

Excitation LIF spectra were measured in the waist region of the excimer beam for dye laser wavelengths varying from 224 to 227 nm (b in Fig. 2). The delay between dye laser and excimer laser pulse was 1 μs. The measured spectrum agrees well with a simulated spectrum of NO at 300 K (a in Fig. 2), which was calculated with LIFBASE.[8] Only some differences in relative line intensities between the experimental and simulated spectrum can be observed. Varying the temperature in the simulations did not improve the correspondence with the experimental data. Even though the NO is unlikely to be formed in thermal equilibrium, it is expected that the NO will have thermalized 1 μs after formation. We attribute the discrepancy to the nonlinear process of NO formation, which results in a strong influence of pulse-to-pulse fluctuations in the excimer laser intensity on the NO LIF signal. In measurements with only a dye laser pulse, no NO signal was observed. This proves that the NO molecules are formed in the waist region of the excimer beam.

The exact mechanism behind the NO formation is not clear yet, but there is circumstantial evidence that ground state oxygen atoms and N_2^+ ions play a role. The energy of the 193 nm excimer laser photons lies within the Schumann–Runge $B^3\Sigma_u^+ \leftarrow X^3\Sigma_g^-$ system of O_2. The B state of O_2 is strongly predissociative, which results in a dissociation of the main fraction of the excited O_2 molecules into O atoms.

Dispersed fluorescence spectra from the waist region of the ArF excimer beam showed bands that could be attributed to transitions in the $B^2\Sigma_u^+ \rightarrow X^2\Sigma_g^+$ system of N_2^+. These results are an indication for a three-photon absorption process in N_2. In Fig. 3a, the important potential energy curves of N_2 that are involved in this process are shown. The energy of the 193-nm photons is indicated by arrows. From Fig. 3a, it is clear that absorption of three ArF photons

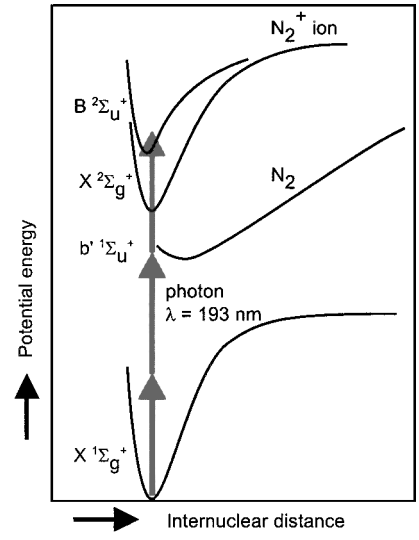

Fig. 3a N_2 potential energy curves; arrows correspond to photons of the ArF laser beam.

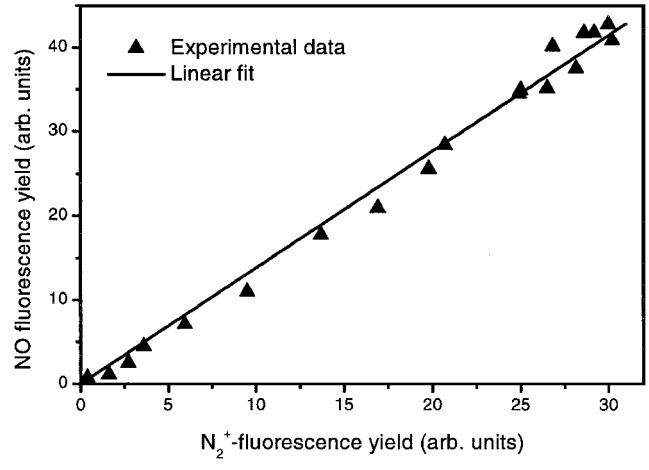

Fig. 3b Relation between the NO fluorescence (226-nm excitation) and the N_2^+ fluorescence in the waist region of the ArF excimer laser beam.

can result in the formation of N_2^+ ions in the B state, which would give rise to the observed fluorescence. The $b'^1\Sigma_u^+$ state might be an intermediate state in the excitation process.

The relation between the N_2^+ and NO formation was investigated as follows. N_2^+ fluorescence of the $B(v'=0) \rightarrow X(v''=0)$ bandhead, induced by the excimer laser, was measured with an optical multichannel analyzer system for different excimer laser powers, on a single-shot basis. NO fluorescence induced by the 226 nm dye laser was measured 11 μs after each excimer laser pulse with the camera system that was described earlier for the tagging experiments. In Fig. 3b the NO LIF signal is plotted against the corresponding N_2^+ signal. The NO fluorescence was measured 11 μs after the excimer laser pulse. The experimental data could be fitted well with a linear dependence, which corroborates the assumption that N_2^+ is an important intermediate species in the formation of NO.

The intensity of the NO LIF signal was measured as a function of the delay between the excimer laser pulse and the dye laser pulse to determine the lifetime of the tagged molecules. In Fig. 4 the NO LIF signal is plotted against the delay time on a logarithmic scale. Experimental data are compared with the function $\log(y) = A * \log(x) + B$. The NO LIF signal was obtained by integrating over the NO emission bands at 237 nm $[A(v'=0) \rightarrow X(v''=1)]$, 248 nm $[A(v'=0) \rightarrow X(v''=2)]$, and 259 nm $[A(v'=0) \rightarrow X(v''=3)]$. The NO signal did not depend on the delay for delay times shorter than 10 μs. For larger delays, the slope of the NO signal decay in Fig. 4 is approximately $-\frac{1}{2}$. This behavior can be explained by diffusion of the formed NO molecules. For a delay smaller than 10 μs

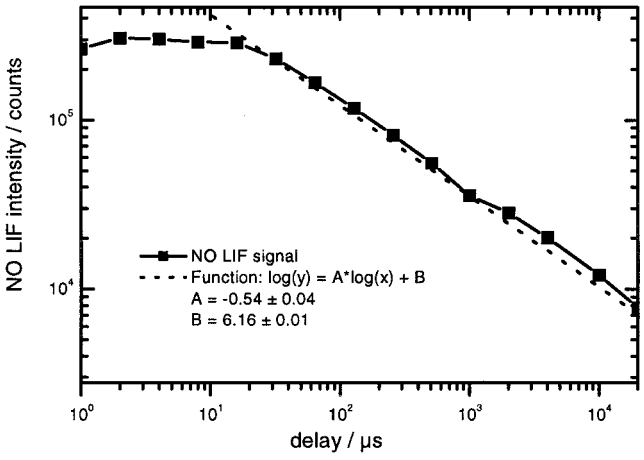

Fig. 4 Integrated NO LIF intensity in the $A(v'=0) \rightarrow X(v''=1)$, $A(v'=0) \rightarrow X(v''=2)$ and $A(v'=0) \rightarrow X(v''=3)$ emission bands as a function of the delay between excimer and dye laser pulse.

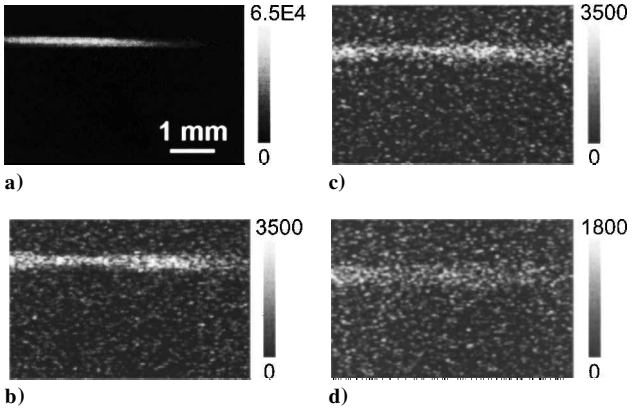

Fig. 5 NO tagging in a laminar airflow with a delay between write and read pulses of a) 1 μs, b) 100 μs, c) 200 μs, and d) 400 μs.

all formed NO molecules are still present within the 0.2-mm-thick dye laser sheet. For larger delays some of the NO molecules have diffused out of the dye laser sheet and are not detected anymore. This diffusion would result in a NO signal that is proportional to $(4Dt + C)^{-1/2}$, with D the diffusion coefficient, t the delay time, and C a constant. Apparently, the lifetime of the NO molecules is so large that it does not noticeably affect the NO LIF signal even for delays in the order of 1–10 ms. After about 1 ms, the NO LIF signal had decreased to 10% of the maximum NO signal.

If we compare the lifetime of the NO molecules to the lifetimes of the tagged molecules in RELIEF, OH tagging, and O_3 tagging, which are on the order of 0.01, 1, and 100 ms, respectively, we can conclude that the lifetime of NO is at least comparable to that of OH and O_3.

Applications

Several flow systems were investigated with the new method. We applied the method to a laminar dry airflow generated by a McKenna-type burner through which only air was flowing, regulated by a calibrated mass flow controller. The excimer laser was focused 3 mm downstream of the flow exit. In Fig. 5, NO LIF images of this flow are shown with a delay between excimer laser and dye laser pulse of 1 (Fig. 5a), 100 (Fig. 5b), 200 (Fig. 5c), and 400 μs (Fig. 5d). The images were obtained with single pulses of both excimer and dye laser. The images were measured with single shots of both write and read laser. From the displacement of the line, a flow velocity of 1.11 ± 0.05 m/s could be derived. The images show a decrease of the NO LIF signal with increasing delay. Furthermore, the width of the line of NO molecules increases as a function of time, also because of diffusion. No deformation of the line can be recognized, which indicates that the laminar flow is not disturbed by the write laser pulse. On the scale of this experiment, therefore, the APART method is not visibly intrusive.

In a steady flow, a number of laser shots can be averaged, which leads to a much better signal-to-noise ratio in the NO LIF images. In the present setup, mean flow velocities in the order of 1 cm/s could be measured this way. In Fig. 6, NO LIF images of the tagged line averaged over 10 (Fig. 6a) and 500 (Fig. 6b) write and read laser pulses are shown. The delay between write and read pulse was 1 μs (Fig. 6a) and 10 ms (Fig. 6b), respectively. After 10 ms, the tagged line had broadened from 0.18 to 1.4 mm by diffusion. The displacement of the line was determined by fitting a Gaussian profile to a perpendicular cross section through the line. In this way the displacement of the line in Fig. 6b with respect to Fig. 6a (0.14 mm) could be determined with an accuracy of 0.02 mm. This gives an average flow velocity of 1.4 ± 0.2 cm/s.

A highly structured flow system we investigated involves an airflow emanating from a pulsed valve (Jordan Company, Grass Valley, California). The valve emits dry air pulses (backing pressure 0.3 MPa) of about 20-μs duration into ambient air through a 0.6-mm-diam flat nozzle. NO is created 0.8 mm below the nozzle exit. NO LIF images were obtained with single laser pulses of both write and read laser. In Fig. 7, the results are presented for a delay of 50 μs (Fig. 7a), 200 μs (Fig. 7b), 400 μs (Fig. 7c), and 1 ms (Fig. 7d) between excimer and dye laser pulses. Again the decrease in NO LIF signal and the broadening of the written profile as a function of the delay time can be recognized. In principle, it would be possible to follow the NO distribution in time by using one excimer laser pulse and several dye laser pulses because the NO molecules are not consumed in the detection process. A dye laser with high repetition rate is necessary for such a measurement.

In Fig. 8, APART images are presented that clearly show the vortex structure that is expected for a pulsed valve flow. Figure 8a

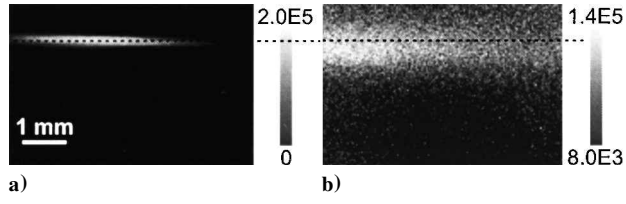

Fig. 6 NO tagging in a laminar airflow; NO LIF images measured after excimer pulse: a) 1 μs and b) 10 ms.

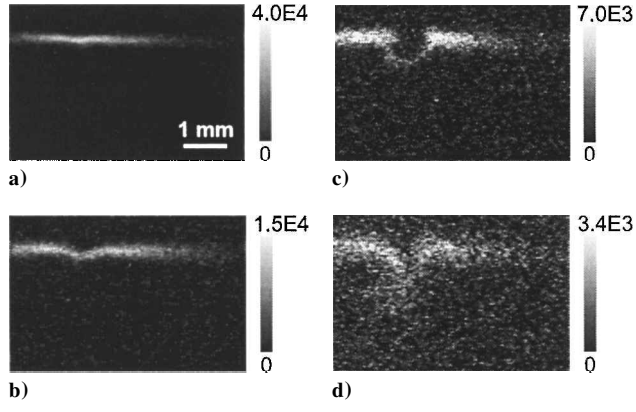

Fig. 7 NO tagging of a pulsed airflow with a delay between write and read pulses of a) 100 μs, b) 200 μs, c) 400 μs, and d) 1 ms.

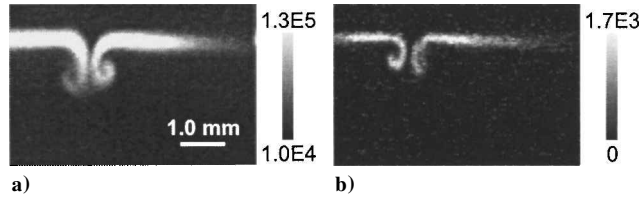

Fig. 8 NO tagging of a pulsed airflow: a) integration over 500 laser pulses (delay 300 μs) and b) single-shot image (delay 100 μs).

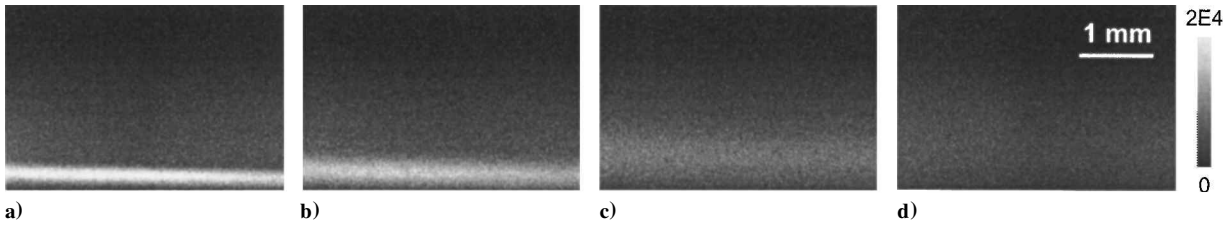

Fig. 9 NO tagging in a flat premixed CH_4/air flame; images were averaged over 100 laser pulses. Delay between excimer and dye laser was a) 1 μs, b) 10 μs and c) 100 μs, and d) measured in the absence of an excimer laser pulse.

was obtained by averaging over 50 laser pulses with a delay of 300 μs between write and read laser pulses. The vortex structure could already be recognized clearly with a single laser shot (Fig. 8b). The experimental conditions in the measurements were comparable to those of Fig. 7; only the maximum valve aperture was larger.

We also applied the APART technique to reactive flows. The results obtained in a flat premixed CH_4/air flame (Fig. 9) indicate that the method can be used to determine velocities in flames. The measurements were performed in the same McKenna-type flat flame burner as was used for the laminar air flow measurements. The excimer laser beam passed over the burner surface at a height of about 0.5 mm. The equivalence ratio ϕ was set to 1.0. In Fig. 9, APART images are shown obtained at a delay of 1 (Fig. 9a), 10 (Fig. 9b), and 100 μs (Fig. 9c) after the excimer laser pulse. Figure 9d was measured in the absence of an excimer laser pulse. Figure 9d shows the NO LIF signal of the NO molecules that were formed in the flame; its distribution is indicative for the dye laser beam profile. All images in Fig. 9 are shown with the same intensity scale and are averaged over 100 write and read pulses. It is clear that the line of NO molecules formed by the excimer laser pulse can very well be separated from the natural NO in the flame. Also, the initially written line is seen to remain undisturbed during advection by the laminar flow. Again, this indicates that the APART method is effectively nonintrusive under the conditions of this experiment. The flow velocity in the flame, again determined from the displacement of the tagged line, amounted to 2.3 ± 0.3 m/s, assuming a uniform temperature in the region in which the tagged NO molecules reside. This value corresponds well with the flow velocity determined by PIV, which was equal to 2.4 m/s (personal communication from K.-J. Boschaart).

This last example demonstrates that APART is a promising technique for nonseeded velocimetry in reactive flows. The only restriction is that the laser-induced NO can be distinguished from the NO that was formed in the combustion process itself. We have shown that this is the case in combustion processes at a relatively low temperature, such as a methane–air flame ($T \approx 1800$ K at this height in the flame).

OH tagging by H_2O photolysis can also be applied in reactive flows. Wehrmeyer et al.[5] determined the velocity in a H_2/N_2–air flame with $T = 2340$ K and obtained a value of 22 m/s with an accuracy of about 3 m/s. This accuracy was obtained with single-shot measurements. If we assume that the signal is shot noise limited, this corresponds to an error of 0.3 m/s after averaging over 100 pulses. This error is comparable to the error in the velocity determined from the APART measurements in a CH_4/air flame after averaging over 100 pulses. More research is necessary to investigate whether APART can also be applied in combustion processes at higher temperatures containing larger amounts of natural NO.

Conclusions

We have shown that APART can be used to measure velocities in both laminar and pulsed airflows. Experimental results were given that indicate the importance of N_2^+ in the NO formation mechanism. The decay of the NO LIF signal intensity appeared to be mainly determined by diffusion of the produced NO molecules out of the dye laser beam. This indicates that the lifetime of the NO molecules is at least 1–10 ms. Velocities could be determined with an accuracy of about 5% with single pulses of both the excimer (write) and the dye (read) laser. After averaging over 500 laser pulses, velocities as low as 1 cm/s could be measured. Furthermore, velocities in a CH_4/air flame could be measured with an accuracy of 13%. These first results indicate that APART is a useful new method for velocimetry in nonseeded airflows which can compete with other techniques such as RELIEF and OH and O_3 tagging.

Acknowledgments

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