Rayleigh Scattering Measurements of the Gas Concentration Field in Turbulent Jets

M. Carla Escoda* and Marshall B. Long†
Yale University, New Haven, Connecticut

A technique based on Rayleigh scattering has been developed to measure the concentration field in a cross section of a turbulent gas jet. Such measurements enable one to quantitatively study turbulent mixing mechanisms and structures. A similar experiment was previously performed using Lorenz/Mie scattered light from aerosol particles introduced into the nozzle gas. The Rayleigh technique provides better spatial resolution, avoiding the limitations due to aerosol seeding, and in particular is capable of monitoring molecular diffusion, a fact which should be of importance in studying reactive turbulence.

Introduction

THE need for high-resolution nonintrusive methods of obtaining quantitative information on mixing in turbulent flows has led to the development of light scattering techniques to determine molecular concentration and temperature profiles. Rayleigh and Raman scattering have proved to be particularly valuable for making measurements at single points or along a line in the flow.[1-6] However, recent interest in large-scale coherent structures in turbulence demonstrates the need for instantaneous two-dimensional measurements. The study of turbulent eddies and their relation to the large vortical structures developed further upstream in the transition region of a jet requires fine temporal and spatial resolution and the simultaneous probing of an entire flow region. In addition, turbulent mixing often takes place in environments that are relatively inaccessible or destructive to conventional probes. Since light scattering methods can monitor events in an essentially "instantaneous" manner and do not require the introduction of physical probes into the flow, they are extremely valuable to the experimental study of turbulence.

A Previous Light Scattering Technique

Our application of Rayleigh scattering to the two-dimensional mapping of concentration in the near field of a freon jet is similar to a technique previously developed to infer the concentration by measuring the Lorenz/Mie scattering from an aerosol-seeded jet.[7]

Both experiments utilized the fact that the intensity of scattered light is proportional to the number of scatterers in the illuminated volume, i.e., to the number of seeded particles per resolution element in the Lorenz/Mie case or to the number of gas molecules per resolution element in the Rayleigh case. The variation of scattered light intensity across a sheet of laser light in a plane of the jet is recorded with a computer-controlled low-light-level TV camera (PAR OMA-2). The scattered light intensity is interpreted as being proportional to the nozzle fluid concentration.

In regions where two gases mix, the intensity of the Rayleigh-scattered signal is proportional to a weighted sum of the nozzle and ambient gas concentrations, the weights being their different cross sections for Rayleigh scattering. If the ratio of nozzle to ambient gas cross section is sufficiently large, one can ignore the contribution of the ambient gas term in solving for the nozzle gas concentration in these regions. In the case of low velocity jets, i.e., under constant pressure and temperature, such as in our experiments, it is not necessary to make this simplification, as one can solve directly for the concentration of each component gas in the mixture.[1,4] In practice, it is desirable to choose two gases with a large difference in cross section, as small changes in number density are more easily detectable.

The image, i.e., the two-dimensional concentration mapping, is digitized at 10,000 points and stored in the computer, thus providing a measure of relative concentrations in two dimensions. Figure 1 shows a typical instantaneous concentration map (using the Lorenz/Mie technique) produced by gating the TV camera on for a short time compared to the estimated time during which detectable downstream translation occurs (10 μs).

Data acquired in this manner must be corrected to account for the background signal, the camera response, and the nonuniformity of the laser sheet. In the first part of this two-step process, a 100×100 "background" frame, taken without the jet on, is subtracted from the data frame. Then a rectangular glass cell holding a dilute fluorescent dye solution is placed in the path of the laser sheet so that a fluorescent sheet is imaged onto the camera face. (Optical filters are used to pass only the fluorescence.) Since the fluorescence signal is constant across the sheet, the actual recorded signal intensities reflect both the nonuniform response of the camera and the intensity distribution in the sheet, which drops off at either end in the direction of the jet axis and peaks in the center due to the Gaussian profile of the incident beam. The background-corrected frame is finally divided by this response frame.

Although the Lorenz/Mie technique affords a high resolution ($200 \times 200 \times 200$ μm per unit resolution volume[7]), it still does not resolve finer structures known to exist in the turbulent regimes studied. Spatial resolution using the Lorenz/Mie technique is limited by marker shot noise which is inversely proportional to the root of the number of seeded aerosols per unit volume. Marker shot noise is evident in Fig. 1 from the variations in intensity between adjacent points in areas where there is no significant concentration gradient. (At this low Reynolds number of 4160, the potential core extends at least partially into the region under observation. Thus, the intensity variations toward the center of the jet in the upstream region of the profile are the result of marker shot noise and cannot represent the actual concentration profile.) In order to minimize marker shot noise, marker density must be high. Overly heavy seeding of the nozzle gas, however, may lead to multiple light scattering, i.e., the rescattering of signal from one aerosol by another before the original signal reaches the camera face. Furthermore, most of the scattering comes

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*Graduate Student, Applied Mechanics.

†Assistant Professor, Applied Mechanics, and Center for Laser Diagnostics.

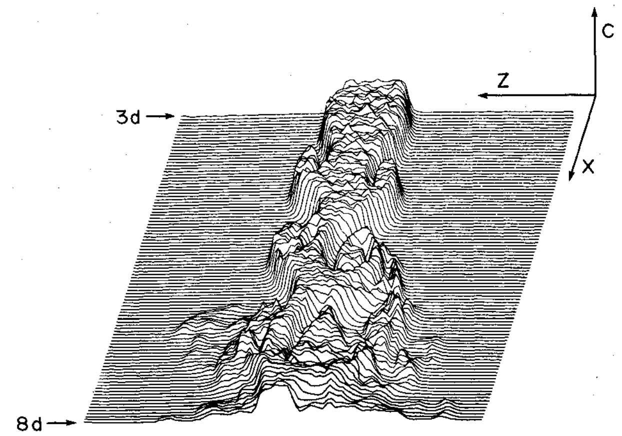

Fig. 1 **Instantaneous concentration profile of a turbulent jet (seeded air exiting into air) obtained by the Lorenz/Mie scattering technique. The nozzle diameter, *d*, was 4 mm and the Reynolds number was 4160.**

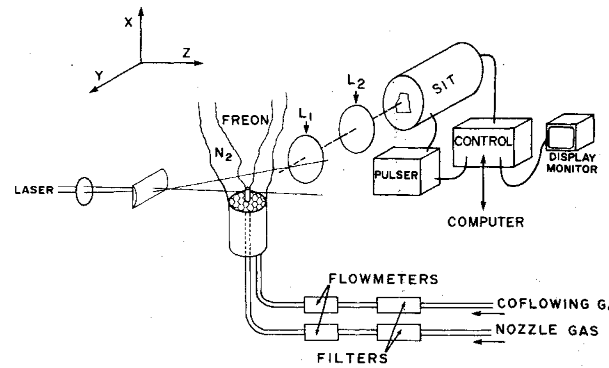

Fig. 2 Experimental configuration for the Rayleigh scattering technique. The inner nozzle has a diameter of 6 mm. Both gas streams are filtered with 0.3-μm filters to remove dust particles from the flow.

from the largest particles so that, in a polydisperse flow, large intensity variations between two scattering volumes under identical flow conditions may arise from a small variation in the number of large aerosols. While in a unit resolution volume measuring $200 \times 200 \times 200$ μm there are approximately 1000 aerosols, an increase in magnification, resulting in a smaller resolution volume, means a decrease in marker density and thus an increase in marker shot noise. In general, therefore, seeding levels restrict resolution in the Lorenz/Mie scattering technique.

The large size of aerosols relative to gas molecules limits their ability to follow flows in which there are large accelerations. Also the instantaneous concentration distributions obtained by the Lorenz/Mie scattering technique could be misleading in regions where molecular diffusion effects are important, since the seeding particles do not diffuse. The Rayleigh experiment was expected to overcome the major noise and resolution limitations of the Lorenz/Mie technique, since scattering from molecules of the nozzle gas was measured rather than that from particles introduced into the flow.

Rayleigh Scattering Technique

Because the intensity of the scattered Rayleigh signal from gas molecules is several orders of magnitude lower than that of the Lorenz/Mie signal from aerosols, the Rayleigh experiment requires higher incident laser power, a "clean" environment to eliminate scattering from particulates, and stray light suppression. In addition, it is desirable to maximize the scattering cross-section ratio between ambient and nozzle gases. This cross section depends on the incident wavelength and the refractive index as follows[8]:

$$\sigma_{R_i} = \frac{4\pi^2}{\lambda_0^4}\left[\frac{n_i - 1}{N_0}\right]^2$$

λ_0 is the incident wavelength, n_i the refractive index, and N_0 the number density. Thus we chose a gas of high refractive index (such as freon) discharging into a gas of much lower index (e.g., air, He) to maximize the contrast between scattered signal intensities from the nozzle and ambient gases. This coflowing configuration consists of a nozzle with a diameter d of 6 mm surrounded by a coaxial honeycomb. To eliminate Lorenz/Mie scattering from dust particles, we cleaned both flows using in-line microfiber filters (pore size = 0.3 μm). The experimental setup (see Fig. 2) was similar to that of the Lorenz/Mie experiment.[7] While an argon ion laser ($\lambda_0 = 488$ nm) provided enough energy (20 μJ in 10 μs) for the Lorenz/Mie experiment, it was necessary to use a pulsed Nd:YAG laser ($\lambda_0 = 532$ nm) that provided 20 mJ in a 15-ns pulse. The laser beam was focused into a sheet approximately 50 μm thick over the jet, and two camera lenses (57 and 135 mm focal lengths) collected and focused the scattered light from a portion of the illuminated sheet onto the face of the camera. The digitized data for each instantaneous shot were stored in an 80×100 array on magnetic tape.

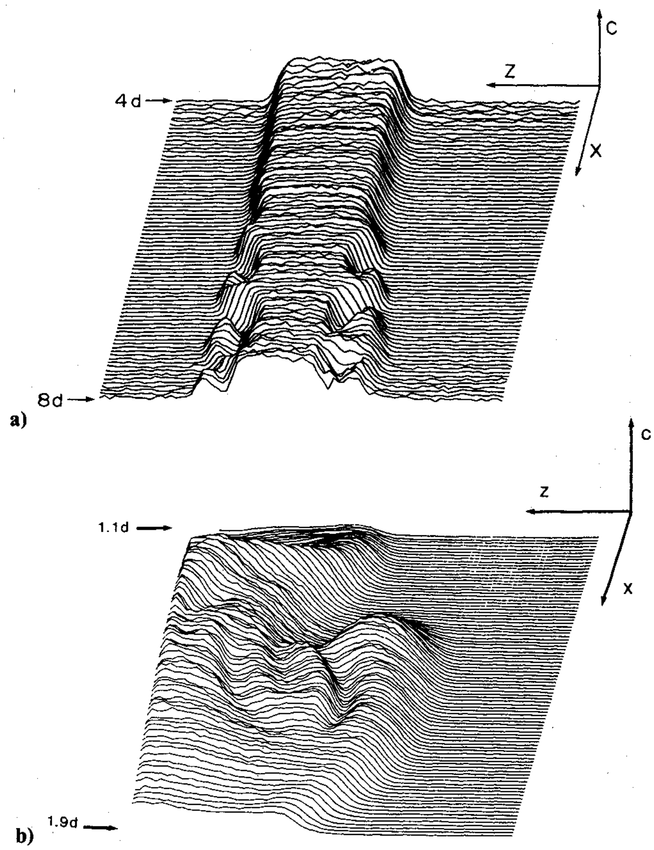

Fig. 3 Instantaneous concentration profiles of a coflowing turbulent jet (freon into nitrogen) obtained by Rayleigh scattering. a) The entire jet (from 4*d* to 8*d*) imaged onto the computer-controlled TV camera. b) A smaller region of the jet imaged onto the camera, giving a spatial resolution of $48 \times 50 \times 50$ μm. In both cases, the freon exit velocity was 4.6 m/s and the velocity ratio, *m*, was 0.18. The density ratio, *n*, of freon and air is 0.20.

Figure 3a shows a typical shot for freon discharging into nitrogen. Note the absence of marker shot noise, as compared to the Lorenz/Mie profile in Fig. 1. The coflowing jets can be characterized by the parameters $m = v_1/v_2$ (velocity ratio) and $n = \rho_1/\rho_2$ (density ratio) where *1* signifies the ambient gas and *2* the nozzle gas.[9] In this case, $m = 0.18$ and $n = 0.20$. A unit resolution volume measures $130 \times 150 \times 50$ μm since the entire area under observation was $1.7d \times 2.5d$. Subsequent magnification was accomplished through switching the positions of the camera lenses and a smaller area of the jet was monitored (see Fig. 3b). The spatial resolution is now $40 \times 48 \times 50$ μm, and structures on the order of 100-200 μm are now clearly resolvable.

In the Rayleigh experiments, data were corrected for camera response and intensity distribution in the sheet by forcing the coflowing gas (in this case, N_2) through both the nozzle and the honeycomb to provide a "constant" intensity field over the entire camera face. The "response" frame was obtained by averaging over several such frames. This correction was less successful than the one made for the Lorenz/Mie data so an additional step was taken. One column, or track, of data was chosen in the center of the data frame, as close as possible to the jet centerline (where presumably the actual intensities were fairly constant in the direction of the jet axis), and divided into each of the remaining tracks. Thus the artificial peak toward the center of each frame due to nonuniform intensity distribution in the sheet was suppressed.

Because of the refractive index difference between nozzle and ambient gases, it is conceivable that small structures in the flow would be optically distorted. Since the characteristics of the interface between the two gases are not known, the optical paths of signals from two adjacent points in the flow may be different, and defocusing would result. This would effectively limit the spatial resolution. To examine the actual effect of this index mismatch, a 10-μm optical fiber was placed perpendicular to the jet axis at the same distance downstream from the nozzle exit as in the Rayleigh experiments. The fiber was illuminated perpendicular to its axis with a lamp and the light scattered off it was focused onto the TV camera. Initially, the image of the fiber was recorded without turning the jet on. With the jet on, and under the same flow conditions as in the experiment, there was no measurable optical distortion of the fiber image. We can therefore conclude that the potential resolution-limiting mechanism arising from the density difference between the ambient and nozzle gases does not affect our experiment down to the limit of our current resolution.

Molecular Diffusion Analysis

That the Rayleigh scattering technique presents a significant improvement over Lorenz/Mie scattering can be demonstrated by its ability to detect molecular diffusion effects. While these effects are insignificant compared to the convection of large-scale structures in nonreactive flows, they become important in reactive flows in which instantaneous (and not mean) concentration profiles determine the local reaction rate. In regimes where molecular diffusion has affected the flow, we do not expect optical diagnostic techniques based on scattering from seeded particles to reveal diffusion-related structure changes. For example, assuming infinite resolution, i.e., no marker shot noise, we expect to see sharp, clearly defined boundaries indicative of the extent to which the aerosol particles have followed the "rolling up" of vortices in the process of entrainment of the ambient gas by the nozzle fluid. In reality, we do not expect these boundaries to be so clearly defined since diffusion of the nozzle fluid must have occurred to smooth out the profile somewhat. Information on this diffusion will be available from the Rayleigh data, provided the diffusion length (i.e., the distance over which the freon has diffused) is greater than the distance between two adjacent resolution volumes.

The distance over which one gas diffuses into another depends on the time interval allowed for diffusion. According to Becker and Massaro,[10] vortices roll up one-half revolution per wavelength of translation. In this case, the wavelength is on the order of the nozzle diameter (see Fig. 3a) and the vortices are convected with a speed approximately equal to the average exit speed of the nozzle and ambient gases or 3 m/s. Therefore, the time required to form a fold in a vortex is (6 mm)/(3 m/s) or 2 ms.

Treating the freon-air mixture as a binary mixture and regarding the molecules as rigid spheres, one can estimate the diffusivity, D, of freon in air to be roughly 0.1 cm^2/s. In a time interval of 2 ms, the freon would therefore have diffused a distance equal to $\sqrt{2D(\Delta t)}$ or $(2 \times 0.1 \times 2 \times 10^{-3})^{1/2} = 200$ μm. Since the spatial resolution in the Rayleigh experiment is on the order of 50 μm, the effects of molecular diffusion should be detectable.

Experimental confirmation is provided by a direct comparison of Lorenz/Mie and Rayleigh scattering under the same flow conditions (low flow rate). Since the flow from the original nozzle could not be seeded, a 4-mm-diam nozzle was used, with freon discharging into air ($m=0$). In the Lorenz/Mie experiments, freon seeded with submicron sized particles was used and, for the Rayleigh case, filtered freon was discharged into the ambient air.

A visual comparison of many instantaneous nozzle gas concentration distributions such as those shown in Figs. 1, 3a, and 3b reveals clearly that, on the average, concentration distributions obtained by the Lorenz/Mie scattering technique have steeper gradients near the jet boundaries than those

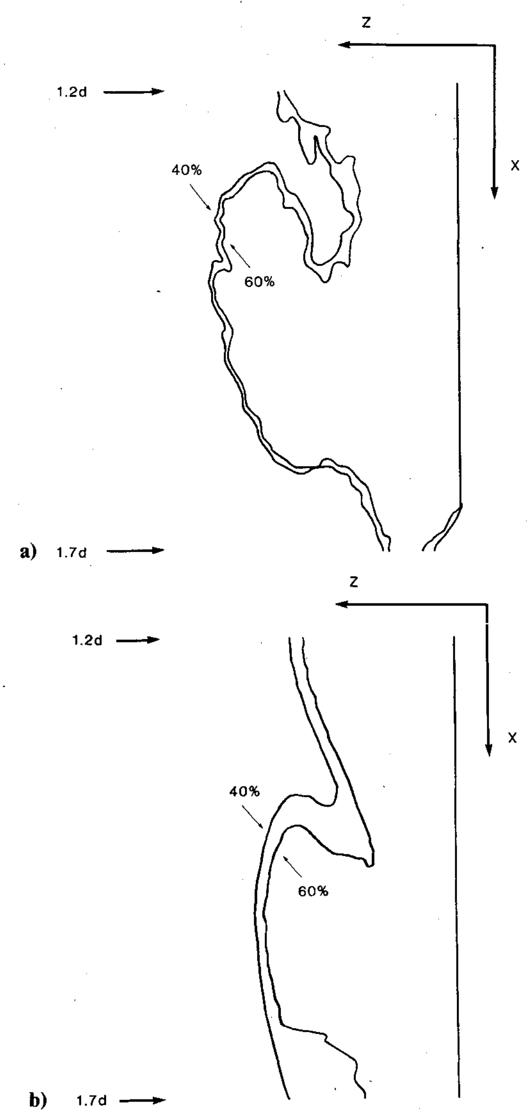

Fig. 4 Comparison of constant concentration contours obtained from a) Lorenz/Mie scattering and b) Rayleigh scattering. The outer curve in each figure represents the portion of the jet where the concentration is 60% of the maximum jet concentration; the inner curve is the 40% contour. The average distance between the contours indicates the steepness of the concentration gradient. The data were for freon exiting into ambient air.

obtained by the Rayleigh scattering technique. The difference in steepness of the concentration profiles is clearly demonstrated in Fig. 4. The two adjacent curves in each case represent two constant concentration contours, which are 40 and 60% of the maximum nozzle gas concentration. The average distance between the two curves is an indication of the steepness of the concentration gradient in that region. This distance is consistently larger in the case of Rayleigh scattering (under the same flow conditions) and is therefore evidence of molecular diffusion.

Conclusion

A technique based on Rayleigh scattering has been developed to measure the instantaneous two-dimensional concentration field in a section of a turbulent gas jet. A similar experiment was previously performed using Lorenz/Mie scattered light from aerosol particles introduced into the nozzle gas. The Rayleigh technique provides finer spatial resolution (on the order of 50 μm), avoiding the limitations due to aerosol seeding density, and in particular is capable of monitoring molecular diffusion.

Because the concentration profile can be directly inferred from the scattered intensity profile of the gas molecules themselves, it should be possible to make measurements in the far field of a jet (more than 12 diameters downstream) and to attain even finer resolution, given sufficient incident laser power. (The required laser power is inversely proportional to the number density of scattering molecules which decreases linearly with distance downstream. Thus, a shift to the far field requires an increase in incident power directly proportional to the distance of the region under observation from the nozzle exit.) With these capabilities, it should become possible to detect small-scale turbulence.

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