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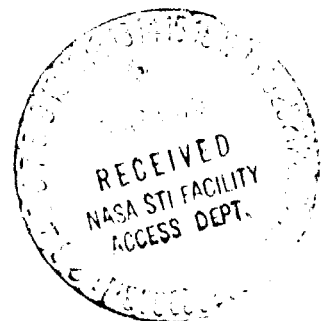
R. L. McKenzie, D. J. Monson, R. J. Exberger

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# **Time-Dependent Local Density Measurements in Unsteady Flows**

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## Time-Dependent Local Density Measurements in Unsteady Flows

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### Abstract

A laser-induced fluorescence technique for measuring the relative time-dependent density fluctuations in unsteady or turbulent flows is demonstrated. Using a 1.5-W continuous-wave  $Kr^+$  laser, measurements have been obtained in 0.1-mm-diameter by 1-mm-long sampling volumes in a Mach 3 flow of  $N_2$  seeded with biacetyl vapor. A signal amplitude resolution of 2% was achieved for a detection frequency bandwidth of 10 kHz. The measurement uncertainty was found to be dominated by noise behaving as photon statistical noise. The practical limits of signal-to-noise ratios have been characterized for a wide range of detection frequency bandwidths that encompasses those of interest in supersonic turbulence measurements.

### Introduction

We describe here a simple and nonintrusive laser-induced fluorescence technique for measuring local density and its

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time-dependent fluctuations in unsteady or turbulent flows. In general, we rely on the fluorescence induced by a high-power krypton-ion laser beam focused into a flow seeded with the organic vapor 2-3 butanedione ( $\text{CH}_3\text{CO}$ )<sub>2</sub>, also known as biacetyl<sup>1</sup> or diacetyl.<sup>2</sup> Biacetyl was first introduced as a flow diagnostic material by Epstein<sup>3</sup> who illuminated seeded flows with a two-dimensional expanded beam from a short-pulse dye laser and photographed the fluorescent sheet to obtain a density field map. In our work, fluorescence from a small sampling volume containing the focal region of a continuous laser beam is viewed photoelectrically to obtain a time-dependent record of the relative density changes. Thus, in the absence of an absolute calibration, this method measures  $\Delta N/\bar{N}$  where  $\bar{N}$  is the time-averaged local density, and  $\Delta N$  is its time-dependent increment. This method is similar in capability to hot-wire probe techniques that measure the relative changes in the product of density and flow speed (i.e., mass flux) but it has the additional advantages of being nonintrusive and deterministic of a single flow variable.

In the past, nonintrusive density measurements have been made most commonly by optical-beam absorption and interference methods.<sup>4,5</sup> More recently, such measurements have been obtained by electron-beam-induced fluorescence.<sup>6</sup> Typically, optical-beam methods provide only a spatially averaged density integrated along the line of sight, and electron-beam techniques are restricted to

low-density flows where electron scattering and fluorescence quenching are not severe. Conversely, the method reported here allows measurements from volumes as small as those sampled by conventional hot-wire probes and with an equivalent time response. The capability of the method to resolve amplitude fluctuations in the signal is limited mainly by signal noise. Thus, unlike electron-beam techniques, this method is best applied at number densities not greatly diminished by flow expansion where the signal is large. If the seed and background gases are mixed in the flow-facility reservoir, adequate sensitivity may be achieved in expansions to low supersonic Mach numbers.

The discussion to follow first characterizes the performance and practical limits of this method in terms of the maximum signal-to-noise ratios achievable for a given detection frequency bandwidth. A demonstration of the technique is then described in which fluorescence and hot-wire signals from a seeded, Mach-3 channel flow of  $N_2$  are compared.

#### Measurement Technique

The experimental arrangement for this technique is illustrated in Fig. 1. The central axes of the flow, the laser beam, and the detector optics all intersect at the optical focal points. The dimension of the sampling volume is determined by the diffraction-limited or aberration-limited diameter of the focused laser beam

and by the axial length of the focal region viewed by the detector through an aperture slit. In our experiments, the beam had a transverse Gaussian profile with a width at half-height of 0.1 mm. It was viewed over an axial length of 1 mm.

The parameters pertinent to this technique are identified by the relation:

$$P_f(t) = P_L \Omega \eta_f (1 - e^{-\sigma N_a(t)L}) \quad (1)$$

where  $P_f(t)$  is the time-dependent fluorescence radiative power (watts) received by the detector;  $P_L$  is the incident laser beam power entering the sampling volume;  $\Omega$  is the collection efficiency of the detector optics;  $\eta_f$  is the fraction of absorbed power that is radiated as fluorescence;  $\sigma$  is the molecular cross section for radiative absorption;  $N_a(t)$  is the time-dependent number density of absorbing molecules in the sampling volume; and  $L$  is the absorption path length observed. In a uniformly mixed gas,  $N_a(t) = X_a N(t)$ , where  $X_a$  is the mole fraction of absorbing molecules and  $N(t)$  is the time-dependent number density of the mixture — the quantity to be measured. When the radiative power absorbed in the sampling volume is sufficiently small compared to  $P_L$ , Eq. (1) can be approximated by a relation linear in  $N$ ; namely,

$$P_f(t) = KN(t) \quad (2)$$

where

$$K = X_a \eta_f \sigma \Omega L P_L \quad (3)$$

is a constant to be fixed and maximized by the experimental arrangement. In the remainder of this section, we discuss the factors contributing to  $K$  and the practical limits determining its maximum value.

The nonadjustable factors in  $K$  that decide the feasibility of the technique are determined by the properties of the seed gas, biacetyl. Biacetyl is unique as a fluorescent seed gas because it combines the advantages of a relatively high vapor pressure, an absorption cross section of usable magnitude at wavelengths accessible with high-power, continuous-wave lasers, and a nontoxic nature. The low toxicity of biacetyl presents no significant hazard but biacetyl liquid is combustible when exposed to an open flame.<sup>2</sup> However, no information has been found that defines the combustion mixture limits for biacetyl vapor mixed with air. Thus, although the possibility exists that biacetyl vapor can be used at low concentrations in air without a combustion hazard, we have used it only in  $N_2$  flows.

The properties of biacetyl that determine the molecular parameter,  $X_a \eta_f \sigma$ , in Eq. (3) are well known.<sup>1,3</sup> Its vapor pressure determines the maximum value of  $X_a$  possible for a given reservoir pressure and temperature. Biacetyl is a liquid at room conditions with a vapor pressure of 40 Torr. Working at room temperature, we have been able to evaporate it into an evacuated reservoir to pressures of 20 Torr without signs of condensation. Subsequent slow



loading of the nitrogen to any final pressure was then possible without further effects of condensation, provided that local unmixed concentrations of biacetyl were not created during the loading process or that an adequate waiting period was allowed to promote re-evaporation. However, even after waiting periods exceeding 1 hr, the variation of our fluorescence signals as a function of initial biacetyl partial pressures became nonlinear for values greater than 20 Torr. Eventual polymerization, which occurred in the biacetyl stored at its vapor pressure, also reduced the evaporation rate and further inhibited the maximum pressure to which it could be loaded.

The radiative properties of biacetyl, as presented by Okabe and Noyes<sup>1</sup> are shown in Fig. 2. Absorption occurs predominantly from a singlet ground state to singlet excited states which then fluoresce in the blue with a conversion efficiency  $\eta_f$  of  $2.5 \times 10^{-3}$  and a radiative lifetime  $\tau_f$  of  $10^{-8}$  sec. Intramolecular energy transfer also channels a large fraction of the absorbed energy to a lower-energy triplet system that then phosphoresces in the green to the ground state. The phosphorescence has a conversion efficiency 60 times greater than fluorescence and a radiative lifetime of  $1.8 \times 10^{-3}$  sec. But, unlike fluorescence, the phosphorescent emission intensity is strongly dependent on temperature, and its conversion efficiency is severely affected by oxygen quenching. Hence, biacetyl phosphorescence is not suitable as an

unambiguous indicator of fluctuating density, particularly in air flows. As an aside, however, the long phosphorescent lifetime in a flowing gas without oxygen causes a luminous streak to appear that is easily visible and provides a well-defined streamline track downstream from the focal region of the laser beam.

The spectral absorption band of biacetyl shown in Fig. 2 includes two principal wavelengths obtainable from commercially available ion lasers. The  $\text{Kr}^+$  line indicated at  $\lambda = 413 \text{ nm}$  is clearly optimum and corresponds to a biacetyl absorption cross-section  $\sigma$  of  $8 \times 10^{-20} \text{ cm}^2$ . At that wavelength, continuous laser powers exceeding 1.5 W may be readily obtained, creating power densities at the focal point approaching  $1 \text{ MW/cm}^2$ . At such power densities, the question of saturation effects on the absorption process for focused laser beams must also be discussed. In a molecular system such as biacetyl, the steady-state saturation power  $P_g$  for an effective area  $A$  at the laser beam focus, is controlled predominately by the fluorescence lifetime. It may be approximated by the simplified relation  $P_g = Ahc/\lambda\sigma\tau_f$ , where  $hc/\lambda$  is the energy of a photon at wavelength  $\lambda$ . Given the parameters above and a laser beam focused to a 10- $\mu\text{m}$ -spot diameter, we compute a saturating beam power  $P_g$  of 470 W. Clearly, this value far exceeds the capabilities of the lasers in use and validates the use of Eq. (2) in this application. Similarly, with the parameters above, one can also compute the power absorbed in the sampling volume from a

1-W laser and find that, at our Mach-3 test condition, local gas heating and its associated effect on local density are also insignificant.

The  $\text{Kr}^+$  laser used in our experiments was a Spectra-Physics Model 171-01 capable of broadband power greater than 1.5 W at several simultaneous wavelengths in the spectral region from 406 to 422 nm. The beam was expanded and refocused through uncoated fused-silica optics and windows in the flow channel. Due to reflection and scattering losses, only 45% of the initial beam power was delivered to the sampling volume.

The detector optics included a Fresnel collection lens with a 25-mm focal length and a 25-mm clear aperture. The light then passed through a band-pass filter combination transmitting wavelengths centered at 460 nm with a full bandwidth of 60 nm. Scattered laser light and phosphorescent emission were blocked to insignificant levels. The laser focal region was imaged with a magnification of 10 on a 1-cm-aperture slit so that light from a beam path of  $L = 1 \text{ mm}$  passed through to the detector. With this arrangement, the total collection efficiency was  $\Omega = 0.015$ .

The apparatus described provides a value of  $K$  within a factor of 2 to 3 of the practical maximum value achievable (we believe) without exceptional improvements in laser power. In the section to follow, we shall show that the measurement uncertainty or amplitude resolution depends on the square root of  $K$  so that a much greater improvement would be necessary to significantly enhance the measurement capability beyond that described here.

### Capabilities and Limitations

The amplitude resolution, or uncertainty, in the measurement of random time-dependent signals characteristic of turbulent flows is determined in this application by the signal-to-noise ratio achieved with a noise-free signal source. In these experiments, the dominant noise source behaves as photon statistical noise (shot noise), a fundamental property of both the fluorescence emission and photodetection processes. All other noise sources, such as electrostatic, electronic thermal, and laser oscillation noise have been reduced to subordinate levels at the detection frequencies of interest here. The signal-to-noise ratio  $S/N$  can then be simply related to the experimental parameters by<sup>7</sup>

$$S/N = \sqrt{\frac{\eta_D}{2h\nu} \frac{K\bar{N}}{\Delta f}} \quad (4)$$

where  $\eta_D$  is the quantum efficiency of the photodetector,  $\Delta f$  is the detection frequency bandwidth,  $h\nu$  is the average energy per photon received by the detector, and  $K\bar{N}$  is the fluorescence power received by the detector, as defined by Eq. (2). However, in this case, the over-bar signifies  $\bar{N}$  as a time-averaged value. The only detector parameter of importance is  $\eta_D$ , regardless of detector gain or detection area. In these experiments, an RCA C31007G photomultiplier was used with a nominal  $\eta_D = 0.19$  at  $\lambda = 460$  nm. Having chosen a detector with a value of  $\eta_D$  close to the largest typically available, the measurement capability can then be

characterized for all circumstances by measuring the  $S/N$  obtained for several  $\Delta f$  at the maximum signal,  $K\bar{N}$ , achievable. The presence of flow is unimportant.

Signal-to-noise measurements were made using the flow channel as a nonflowing vessel filled with biacetyl vapor to pressures up to 20 Torr. The results are illustrated in Fig. 3. In some cases, air was added as a background gas to pressures of 520 Torr to test the effects of oxygen quenching and to inhibit the diffusion of fluorescent gas out of the laser focal volume; however, no significant effect on  $S/N$  was found. Bandwidth was limited by a 6 dB/octave tunable low-pass filter in the recording electronics. The laser was operated at powers up to 1.5 W. The solid line in Fig. 3 was placed by fitting a line with slope 0.5 to the  $\Delta f = 100$  kHz data, thereby verifying the relation of  $S/N$  to  $K\bar{N}$  predicted by Eq. (4) and, hence, the apparent nature of the noise. The dashed lines for other bandwidths were then computed using the  $\Delta f = 100$  kHz data as a reference.

The maximum values of  $S/N$  shown for each  $\Delta f$  in Fig. 3 are limited by the value of  $K$  and represent the limits of our experimental arrangement. With flow,  $\bar{N}$  will be reduced in proportion to the density expansion ratio from the reservoir, thus reducing  $S/N$  accordingly. From Fig. 3, one can expect  $S/N \approx 300$  to be achievable using narrow-band detection at a band pass less than 1 kHz. This implies an amplitude resolution,  $(S/N)^{-1}$ , of 0.3%.

Broadband detection at 100 kHz degrades the signal quality to  $S/N \approx 25$ , giving an implied signal uncertainty of 4%. Although significant improvements are thought only to be possible through large increases in incident laser power, these results appear to be within the useful range for turbulent flow research.

#### Application in a Supersonic Flow

As an initial demonstration of the laser-fluorescence technique, we have used it to measure the density fluctuations in a Mach-3 channel flow. The channel was driven by a 10-m Ludwig tube filled with 20-Torr biacetyl and 520-Torr  $N_2$ . Quasi-steady flow was obtained for 50 msec. Fluorescence measurements were made in the center of the 2-cm-high by 7.5-cm-wide channel at an axial station 5 mm upstream of a small hot-wire probe. Although the hot-wire responds to mass flux, calculations show that for the present test conditions, its signal is dominated by density changes.<sup>8</sup> Thus, the two signals should be directly comparable. An example of the simultaneous record from both signals is shown in Fig. 4. The density perturbations are believed due to weak unsteady shock waves, visible in Schlieren photographs, interacting with the boundary layer. Both signals were filtered identically with 10 kHz low-pass filters. From the results of Fig. 3 and from the average fluorescence signal level, the fluorescence  $S/N$  for conditions of Fig. 4 is about 50, giving an unambiguous amplitude

resolution of at least 2%. The maximum fluctuations shown in Fig. 4 are about 4% of the time-averaged value and occur with a nearly identical waveform in both records. Slight differences may be attributed to changes occurring in the flow between the two sampling volumes and to fluorescence noise. Similar fluctuations were seen in either probe signal in the absence of the other probe, suggesting that no interaction is present. Thus, we interpret the comparison in Fig. 4 to be an adequate validation of the laser-fluorescence technique for measuring relative time-dependent densities in unsteady or turbulent flows of current interest.

#### Concluding Remarks

We have demonstrated a laser-induced fluorescence technique for measuring the relative time-dependent density fluctuations in unsteady or turbulent flows. Using a 1.5-W, continuous-wave  $\text{Kr}^+$  laser, measurements have been obtained in 0.1-mm-diameter by 1-mm-long sampling volumes in a Mach-3 flow of  $\text{N}_2$ . A signal amplitude resolution of 2% was achieved for a detection frequency bandwidth of 10 kHz. The measurement uncertainty was found to be dominated by noise behaving as photon statistical noise. The practical limits of signal-to-noise ratios have been characterized for a wide range of detection frequency bandwidths that encompasses those of interest in supersonic turbulence measurements. For example, data

extend to conditions showing that narrow-band detection at a band-pass of less than 1 kHz allows signal-to-noise ratios up to 300, implying an amplitude resolution of 0.3%. Broadband detection at a bandwidth of 100 kHz degrades the measurement to a maximum signal-to-noise ratio of 25, giving an implied uncertainty of 4%. Significant improvements are thought only to be possible by large increases in incident laser power. The application of pulsed lasers with high pulse energies and statistical data analysis could make such improvements practical.

#### References

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<sup>3</sup>Epstein, A. H., "Quantitative Density Visualization in a Transonic Compressor Rotor," Journal of Engineering for Power, Vol. 99, July 1977, pp. 460-475.

<sup>4</sup>Ladenburg, R. W., ed., Physical Measurements in Gas Dynamics and Combustion, Part 1, Vol. 9, High Speed Aerodynamics and Jet Propulsion, Princeton University Press, 1954, pp. 3-107.

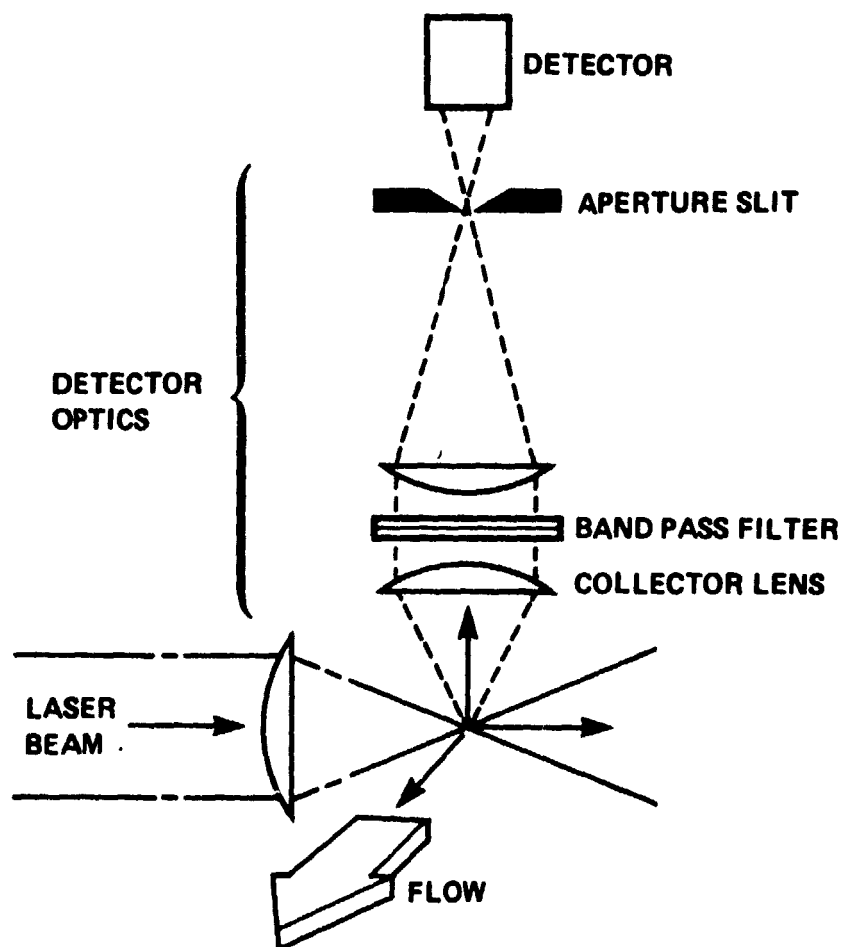
<sup>5</sup>Goulard, R., "Optical Measurements of Thermodynamic Properties in Flow Fields, A Review," AGARD Conference Proceedings, No. 193 on Applications of Non-Intrusive Instrumentation in Fluid Flow Research, AGARD-CP-193, May 1976, paper 13.



<sup>6</sup>Smith, J. A., and Driscoll, J. F., "The Electron Beam Fluorescence Technique Applied to Hypersonic Turbulent Flows," AGARD Conference Proceedings No. 193 on Applications of Non-Intrusive Instrumentation in Fluid Flow Research, AGARD-CP-193, May 1976, paper 16.

<sup>7</sup>"RCA Photomultiplier Manual," RCA Technical Series, PT-61, 1970, pp. 56-76.

<sup>8</sup>Horstman, C. C. and Rose, W. C., "Hot-Wire Anemometry in Transonic Flow," AIAA Journal, Vol. 15, Mar. 1977, pp. 395-401.



**Fig. 1** Experimental arrangement.

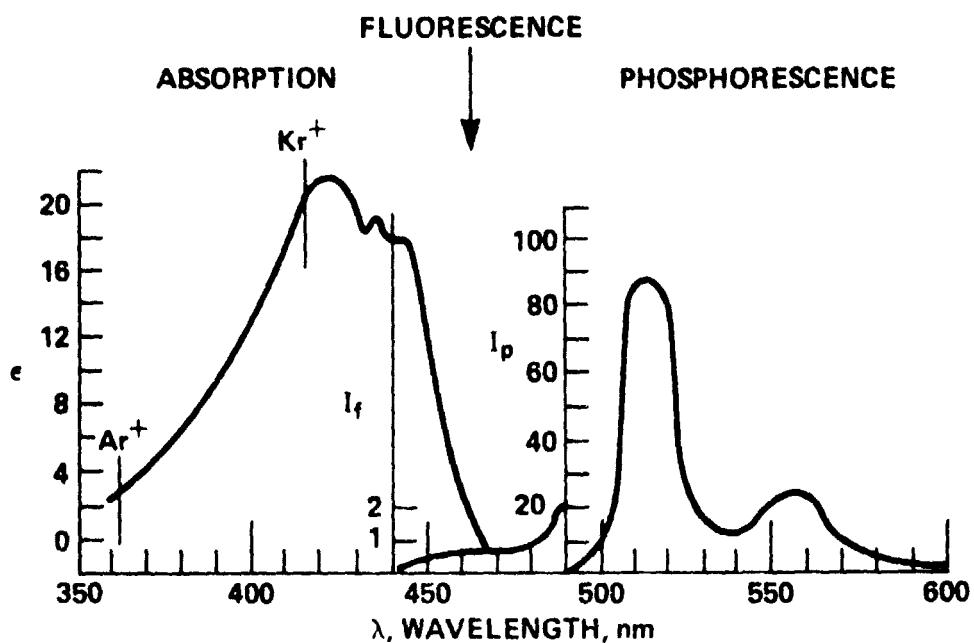


Fig. 2 Absorption and emission spectra of biacetyl. The magnitude of absorption is measured by the extinction coefficient,  $\epsilon$ , as presented in Ref. 1. It may be related to the cross section,  $\sigma$ , in Eq. (1) by  $\sigma(\text{cm}^2) = 3.82 \times 10^{-21} \epsilon$ .  $I_f$  and  $I_p$  are relative emission intensities at low pressure and room temperature for fluorescence and phosphorescence, respectively. Note the scale differences.

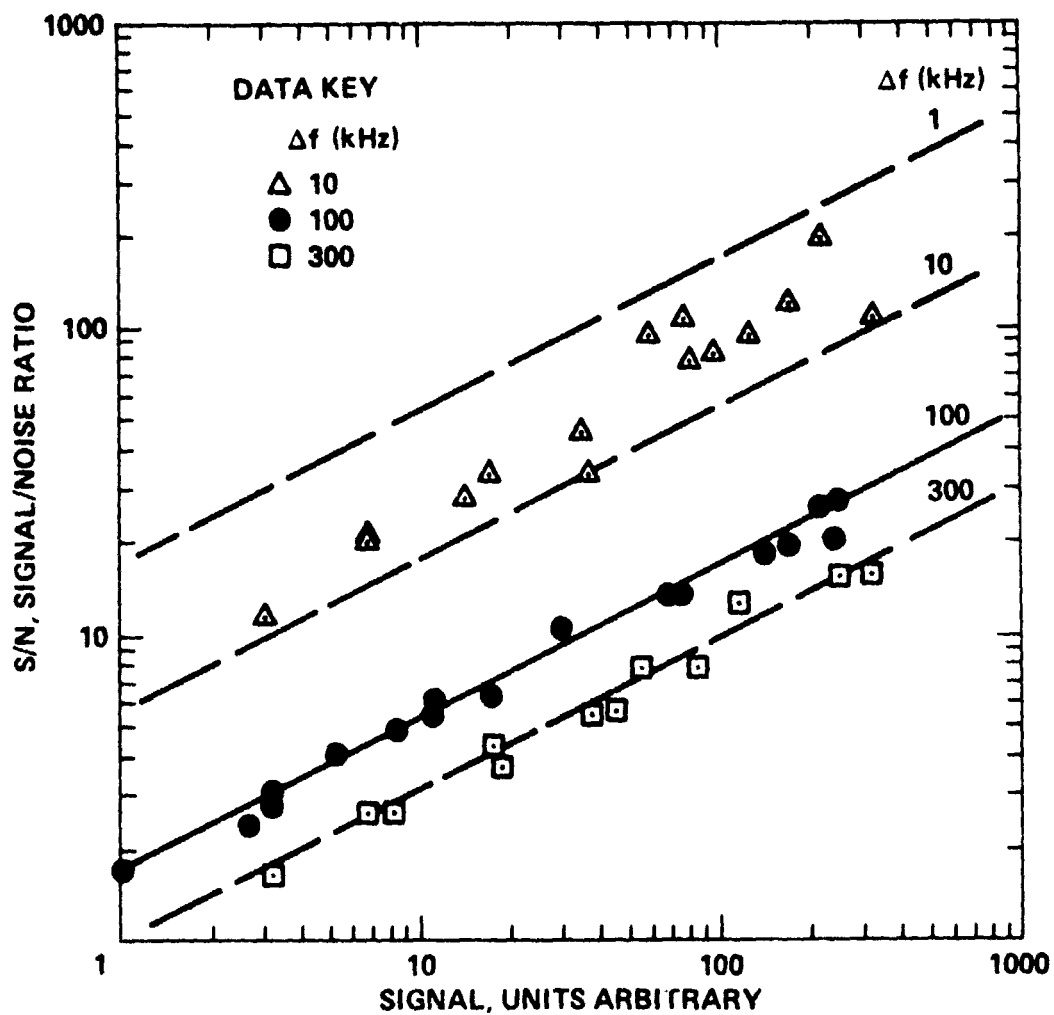


Fig. 3 Signal-to-noise ratios for the detection of biacetyl fluorescence obtained with our apparatus and a comparison with the trends predicted by Eq. (4). The units and magnitude of signal depend on the choice and gain of the photodetection electronics;  $\Delta f$  is the detection frequency bandwidth.

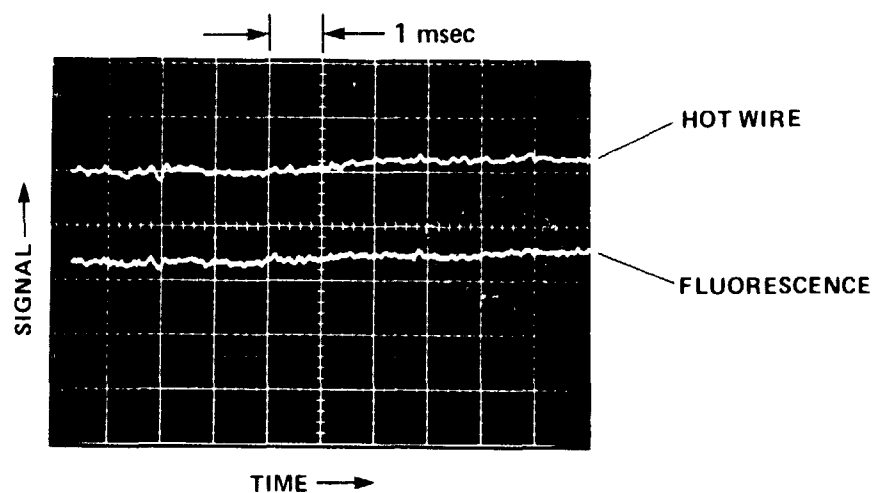


Fig. 4 Comparison of hot-wire and laser-induced fluorescence signals in a Mach-3 channel flow of  $N_2$ . The detection frequency bandwidth was 10 kHz for both signals.