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# Single-pulse gas thermometry at low temperatures using two-photon laser-induced fluorescence in $NO/N_2$ mixtures

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A technique suitable for measuring fluctuating temperatures in supersonic turbulent flows of $N_2$ seeded with NO has been demonstrated in a nonflowing cell. The method relies on the two-photon excitation of two selected ro-vibronic transitions in the NO $\gamma(A^2\Sigma^+, v' = 0 \leftarrow X^2\Pi, v'' = 0)$ band. Measurements between 155-295 K in $N_2$ mixtures containing 300 ppm NO were obtained with single-pulse errors below 4% rms.

In this letter, we describe the demonstration of a laser-induced fluorescence (LIF) technique that provides a practical means of nonintrusively measuring the fluctuating temperatures in low-temperature turbulent wind-tunnel flows. The method, described in detail previously,[1] requires that the flow be seeded with a low concentration of NO, and relies on the two-photon excitation of two selected ro-vibronic transitions in the NO $\gamma(A^2\Sigma^+, v' = 0 \leftarrow X^2\Pi, v'' = 0)$ band. The subsequent broad-band fluorescence energies from each excitation laser pulse are then recorded by digitizing and integrating their time-dependent waveforms. Their ratio is compared with similar data obtained from a reference cell at a known temperature and related to the rotational temperature of the ground-state molecule. As a prelude to its wind-tunnel application, the technique has been demonstrated using a nonflowing cooled cell at pressures and temperatures of interest in a wind-tunnel environment.

There are several features of this method that make it especially suitable for measuring fluctuating temperatures in turbulent flows. These features may be compared with the capabilities of other LIF techniques[2,3] or coherent anti-Stokes Raman scattering (CARS) methods[4] that have all achieved similar accuracies when measuring average temperature, but that are less applicable to single-pulse thermometry in a turbulent environment.

One important feature is the use of a reference cell at known conditions to normalize the effects of pulse-to-pulse variations in laser intensity and frequency distribution. The chemical stability of NO at room conditions makes the use of a reference cell more practical than it would be using other fluorescent species reported, such as OH. Residual noise due to laser and transition bandwidth variations then remains large following normalization only if either of the two lasers is detuned from the center frequency of the transition being excited, and transition broadening is substantially different in the two cells.

Another favorable feature is the relatively small rotational constant of NO compared to hydrogen-bearing molecules. Consequently, upper-level quenching rates and fluorescence lifetimes are fairly insensitive to rotational quantum number and may be assumed equal for all rotational transitions being observed. The use of a reference cell, coupled with the assumption that all rotational states decay radiatively at equal rates, eliminates the need to know the decay rates when computing a temperature from the fluorescence data.

Finally, while rotational CARS has also been demonstrated as a single-pulse technique for measuring low temperatures,[4] it requires a crossed-beam configuration to obtain adequate spatial resolution. Hence, in a turbulent environment where beam steering effects are significant, CARS measurements are expected to contain additional noise components not present in this or other LIF methods.

The experimental arrangement is illustrated in Fig. 1. Two grating-tuned dye lasers are simultaneously pumped at 10 Hz by the third harmonic output of a Quanta-Ray, DCR-1A, Nd:YAG laser. An optical delay line consisting of two 7.5-cm × 12.5-cm dielectric-coated plane mirrors placed 2.5 m apart, delayed a portion of the 355-nm pump beam to the second dye laser, giving a temporal separation of 110 nsec between dye laser pulses. The maximum pulse energy of each dye laser was 5 mJ at 450 nm, in a 5-nsec pulse. The average linewidth was 0.2-0.3 $cm^{-1}$. Amplified spontaneous emission was reduced to less than 5% by spectral and spatial filtering using a direct-vision prism.[5]

The first and second dye laser pulses were tuned to the $J'' = 19\ 1/2$ and $J'' = 7\ 1/2$, $S_{11} + R_{21}$, two-photon transitions, respectively, in the NO $\gamma(0,0)$ band. The two beams were orthogonally polarized, combined collinearly, focused

by a common lens (f = 50 cm), and partitioned into the sample and reference legs by a half-wave plate and a dielectric beamsplitter. Focal spot size was about 0.5 mm. Nearly identical laser energies were directed into the sample and reference cells by placing attenuation plates in one beam path. This was done specifically to match Stark-broadening effects.

The low-temperature sample cell was made of double-jacketed quartz and was cooled by flushing the outer jacket with cold $N_2$ gas from a liquid $N_2$ source. A thermocouple was placed within a few millimeters of the laser beam observation region inside the cell. Nitric oxide concentration for the sample was limited to 300 ppm in 0.5 atm of $N_2$ to match practical wind-tunnel operating limits. The reference cell was at room temperature with an NO concentration of 1200 ppm in 0.5 atm. $N_2$ to minimize photon-statistical noise in the reference signals, while avoiding excessive NO self-quenching.

The broad-band fluorescence from each cell was collected with f/1 fused silica optics, nominally filtered with UV transmitting short-wave pass filters, and imaged through an aperture that limited observation to a 1-mm path length centered on the focal point. Fluorescence waveforms were recorded by solar-blind photomultipliers sensitive in the range 225 to 330 nm and recorded by Tektronix 7912AD 9-bit digitizers interfaced to an HP-1000 computer. The maximum pulse rate for data acquisition was limited to 5 Hz by the data clearing rate from one of the Tektronix units although 12 Hz would have been possible otherwise. Waveform data sets of up to 200 pulses could be stored for each run.

As described in Ref. 1, the data analysis leading to a temperature value for each laser shot requires the ratio of broad-band fluorescence energies resulting from each excitation. The ratio is obtained from each double-pulse waveform by fitting it with a six-parameter function derived for exponentially decaying fluorescence driven by an excitation pulse with a Gaussian temporal profile. The

fit is performed using a nonlinear least-squares method based on a first-order gradient expansion.[6] The two pulses in each waveform are then deconvolved and their individual integrals computed. The integral of each pulse is assumed to be linearly proportional to the total fluorescence energy resulting solely from its corresponding laser excitation with account taken of the laser bandwidth and all collision-broadened transitions falling within the excitation bandwidth.

Figure 2 shows an overlay of the experimental waveforms and their functional fits. In each example, the first pulse results from excitation of the J" = 19 1/2 transition using 1.5-2.0 mJ of laser energy. The second pulse is from excitation of the J" = 7 1/2 transition using approximately 0.5 mJ. Under these conditions, the noise seen in the waveforms is due principally to photon statistics with signal-to-noise ratios in the range of 25-50 for the sample cell (Fig. 2a), and 50-100 for the reference cell (Fig. 2b).

The average fluorescence energy per pulse and corresponding signal-to-noise ratio are shown in Fig. 3 as they vary with laser pulse energy for the $S_{11} + R_{21}$ (J" = 7 1/2) excitation. Conditions are representative of typical wind-tunnel flow. The data are scaled to fall on a theoretical curve calculated using a measured absolute two-photon cross section.[7] Photon-statistical signal-to-noise ratios greater than 100 are implied in the observed fluorescence waveforms. However, a slight departure of the experimental data from the square-law for two-photon absorption occurs well before the expected onset of saturation indicated by the theoretical curve. The effect resembles saturation, but it can be shown as due to resonant Stark broadening by the incident laser field. The broadening begins to dominate the spectral profile at laser power levels of a few hundred megawatts per square centimeter. The large Stark effect is presently believed to be due to near-resonant upward transitions

from the $A^2\Sigma^+$ state that lead subsequently to ionization.[8] It becomes the principal factor affecting the accuracy of the temperature measurement because it limits the maximum laser pulse energy that can be used effectively. Although one can minimize the Stark effects by reducing the power density in the focal volume using a longer focal length lens, it is done at the expense of greater photon-statistical noise in the fluorescence signals. The broadening effects can also be normalized to a significant extent by balancing the partitioning of laser energies into each cell to provide equal power densities.

The results of temperature measurements over the 155-295 K range are shown in Fig. 4, which compares the spectroscopic rotational temperature and thermocouple temperature. The average temperatures calculated from 50 laser pulses for each run are represented by the circles. The error bars indicate the corresponding RMS deviation from each average value. Generally, the averaged temperatures agreed with the thermocouple to within ±2%, while the single-shot temperatures varied between 2.5-4.0% RMS for each data run, depending upon the frequency and power stability of the dye lasers.

The effectiveness of using a reference cell to normalize the fluctuating fluorescence energy ratios was limited by several sources of noise. For the conditions of Fig. 4, it provided only a factor of 2 improvement in the RMS fluctuations compared to unnormalized data. A fundamental limitation is the inherent Poisson noise in the fluorescence detection process, made large by low NO concentrations in the sample cell and by the required use of low laser power densities to minimize Stark-broadening. Increasing the NO concentration by a factor of 4 produced nearly a factor of 2 improvement in the normalizing capability of the reference cell. Another noise source appears to be caused by shot-to-shot laser frequency variations and power fluctuations. They also lead to a degradation of the reference cell normalizing ability because of

dissimilar spectral linewidths which are due to differences in density, temperature, and residual Stark broadening. This effect was verified by using one cell and viewing the same excitation volume with two independent detection systems. There, with the linewidths and Stark effects exactly equal in both channels, and with a high NO concentration to minimize photon noise, normalization to less than 1% in temperature was achieved.

In principle, single-shot density measurements can also be made simultaneously with the temperature measurements for $NO/N_2$ mixtures since NO $A^2\Sigma^+(v' = 0)$ state quenching by $N_2$ is small.[9] However, the measurement of density places greater demands on the measurement accuracy of the fluorescence signals, since density determination is more sensitive to signal noise than temperature. Presently, the noise in the density measurement using 300 ppm NO in 0.5 atm. $N_2$ is about a factor of 2-3 greater than the noise in the temperature measurement.

Improvements in overall measurement capability are anticipated by using a reference cell with temperature and density close to the average wind-tunnel condition being measured, and by narrowing the dye laser bandwidths and maintaining their frequencies close to transition center using intracavity etalons. However, even the present capability is sufficient to provide adequate signal-to-noise ratios for measurements of fluctuating temperatures in supersonic turbulent flows of aerodynamic interest.

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## Figures

Fig. 1. Schematic of the experimental arrangement: BCP, beam-combining polarizer cube; WP, half-wave plate; L, lens; BS, beamsplitter; M, mirror; AP, attenuation plates; COLL, collection system; PMT, photomultiplier.

Fig. 2. Dual-pulse fluorescence waveform (solid line) and computer-fitted function (dashed line): (a) cold sample cell with 300 ppm NO in 0.5 atm $N_2$; (b) room-temperature reference cell with 1200 ppm NO in 0.5 atm. $N_2$. First pulse is fluorescence from the $S_{11} + R_{21}(19\ 1/2)$ excitation; second pulse is from the $S_{11} + R_{21}(7\ 1/2)$ excitation. Vertical amplifier bandwidth, 20 MHz; sweep, 50 nsec/div.

Fig. 3. Average fluorescence signal per pulse as a function of laser energy. The solid line is a calculated curve based on a measured two-photon absorption cross section and the circles are experimental data for the $S_{11} + R_{21}(7\ 1/2)$ excitation of 200 ppm NO in 0.5 atm. $N_2$ at 295 K, with an observed focal volume of 0.3 mm × 1.0 mm.

Fig. 4. Comparison of measured rotational temperature and thermocouple temperature. Cell mixture: 300 ppm NO in 0.5 atm. $N_2$.

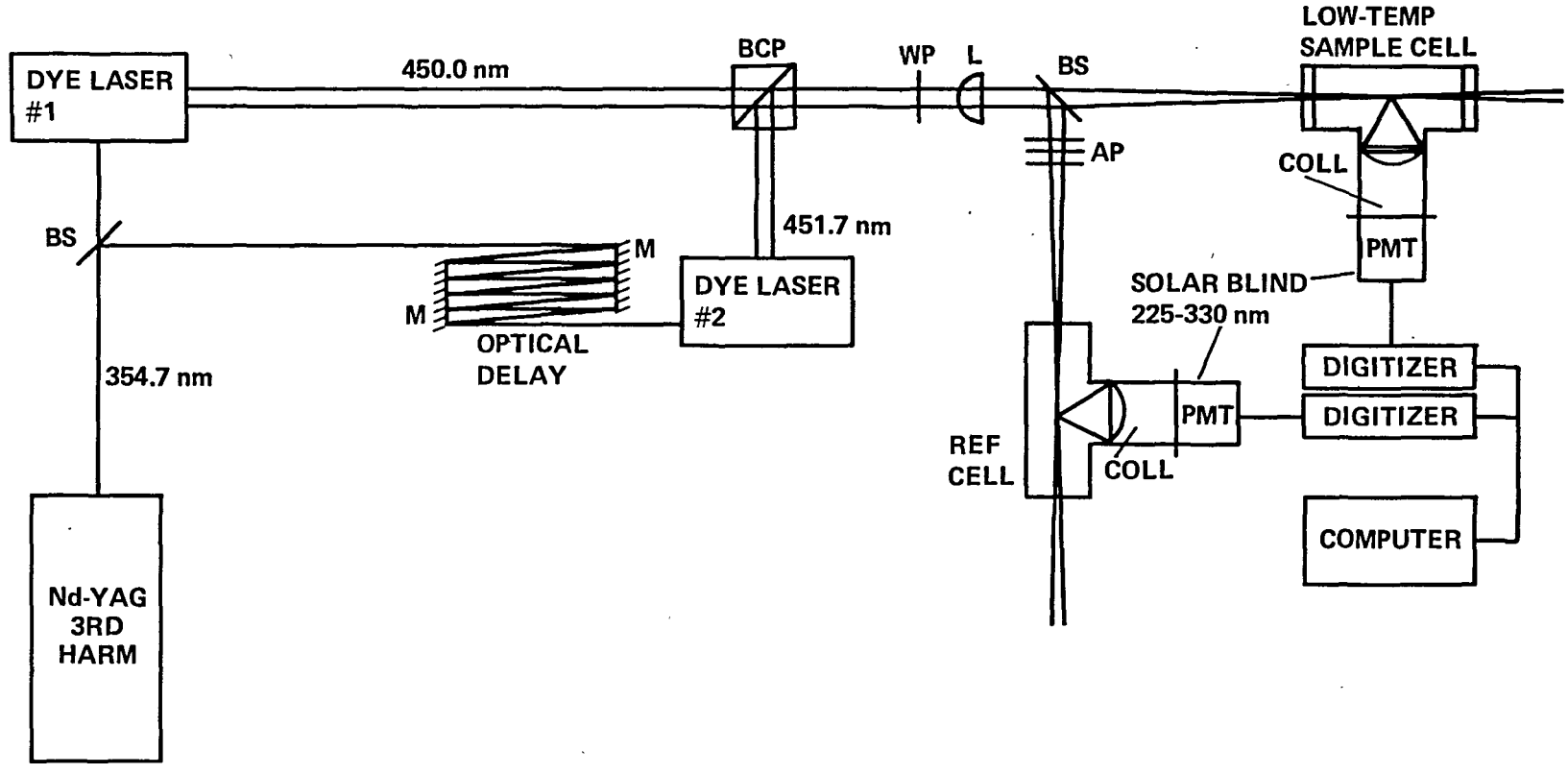
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Figure 1

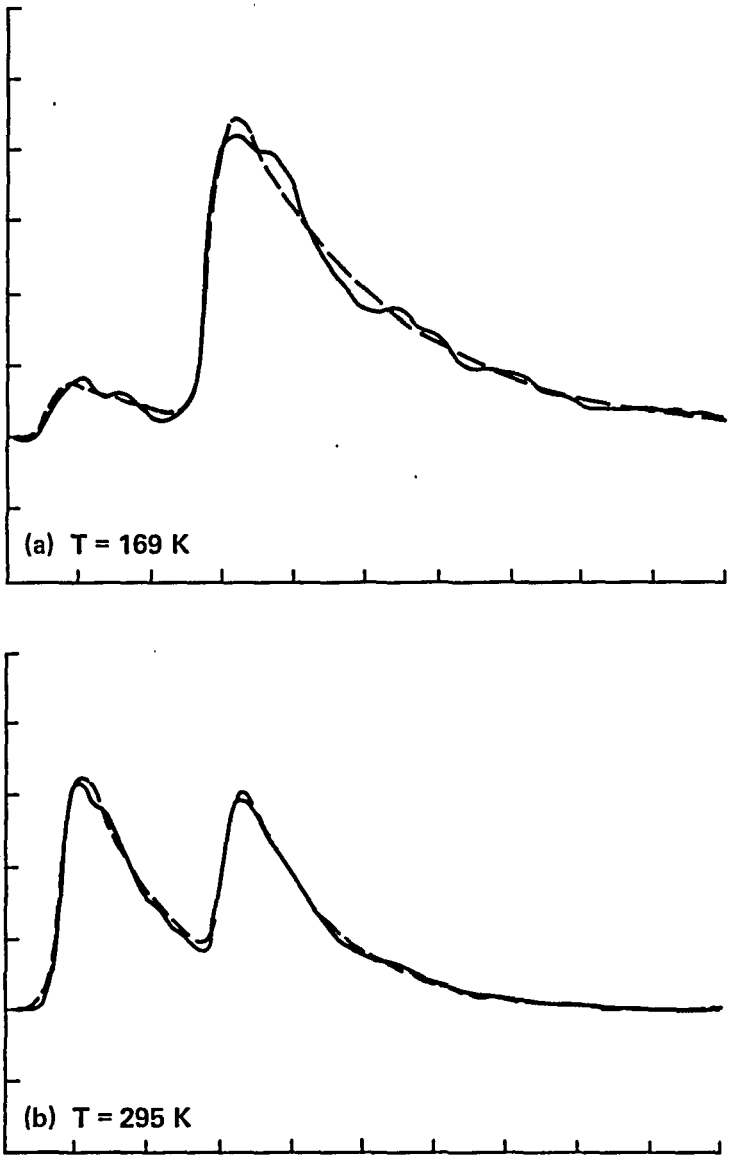


Figure 2

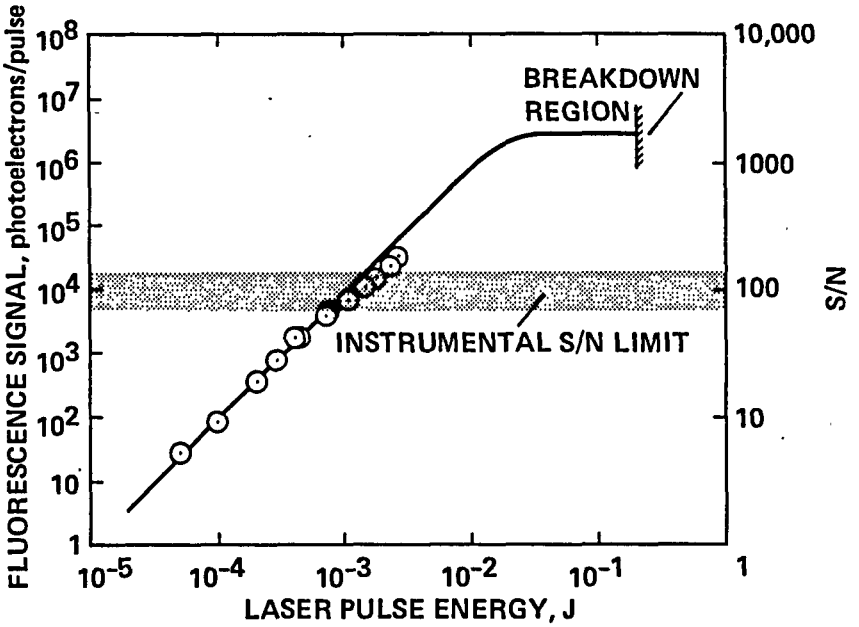
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Figure 3

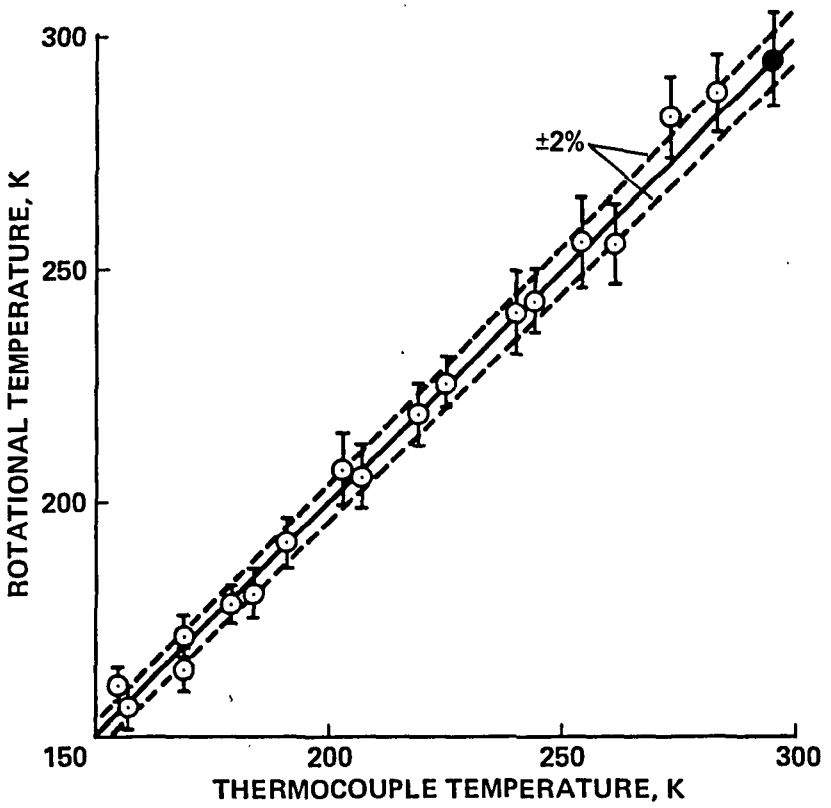
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Figure 4

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15. Supplementary Notes

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16. Abstract

A technique suitable for measuring fluctuating temperatures in supersonic turbulent flows of $N_2$ seeded with NO has been demonstrated in a nonflowing cell. The method relies on the two-photon excitation of two selected ro-vibronic transitions in the NO $\gamma(A^2\Sigma^+, v' = 0 \leftarrow X^2\Pi, v'' = 0)$ band. Measurements between 155-295 K in $N_2$ mixtures containing 300 ppm NO were obtained with single-pulse errors below 4% rms.

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