Two-dimensional imaging of molecular hydrogen in H₂–air diffusion flames using two-photon laser-induced fluorescence

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We report the use of a tunable ArF laser at 193.26 nm to record simultaneous single-laser-shot, planar images of molecular hydrogen and hot oxygen in a turbulent H₂–air diffusion flame. Excitation spectra of fuel and oxidant-rich flame zones confirm a partial overlap of the two-photon H₂ (6,0) Q(11) E, F → X and single-photon O₂ (19,2) P(17) F → X (Schumann–Runge) absorption bands. UV Rayleigh scattering images of flame structure and estimated detection limits for the H₂ two-photon imaging are also presented.

We report the use of a tunable ArF laser at 0.193 μm to record instantaneous planar two-photon laser-induced fluorescence (LIF) images of molecular hydrogen in a turbulent H₂–O₂ diffusion flame. To our knowledge, this represents the first time that molecular hydrogen has been imaged by any method. In addition, partial overlap of the H₂ excitation wavelength with a strong hot-band absorption allows for simultaneous imaging of high-temperature O₂ by using a single laser frequency. This is potentially significant for studies of mixing in turbulent H₂–air combustion, such as in supersonic combustion ramjet engines.

The H₂ excitation/fluorescence scheme, illustrated in Fig. 1, is that first described by Kligler and Rhodes. The two-photon excitation is from the ν' = 0, J' = 1 level of the X ^1Σ₂⁺ state to the ν'' = 6, J'' = 1 level of the E, F ^1Σ₂⁺ state. The two-photon energy is 103 487 cm⁻¹ corresponding to an ArF laser wavelength of 193.261 nm. Note that there is some confusion concerning the labeling of the vibrational levels for the E, F state. We adopt the notation of Huo and Jaffe, who conclude, based on the number of nodes in the vibrational wave function, that the level labeled ν' = 2 by Kligler and Rhodes is more correctly labeled ν' = 6.

The observed fluorescence is from the ν'' = 6 E, F ^1Σ₂⁺ state to the ν" = 1 and ν" = 0 levels of the B ^1Σ₀⁺ state at 830 and 750 nm, respectively. The O₂ hot-band absorption is from the (10,2) R(17) B ← X Schumann–Runge bands at 51 744.9 cm⁻¹ (193.256 nm). The Schumann–Runge fluorescence is in the 200–400-nm range and is easily separable from the near-IR H₂ fluorescence by simple colored-glass filters. This allows instantaneous, separate H₂ and O₂ images to be obtained by using either split-screen techniques or dual cameras, if desired. The two-photon planar LIF experimental arrangement is similar to that used for single-photon fluorescence imaging. An ~10-mJ pulse from a tunable ArF laser (Lambda Physik model 150-EMG) is focused with a 400-mm cylindrical lens into an approximately 10 mm × 100 μm sheet. The waist is at the center of a simple H₂–air diffusion burner constructed by flowing H₂ through a 1.5-mm

Fig. 1. H₂ energy-level diagrams for the relevant states for two-photon LIF.

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asymmetry in the \( \text{O}_2 \) fluorescence intensity. Averaged images of spontaneous visible flame luminosity also displayed this asymmetry, which we conclude is due to imperfections in our burner and not to absorption of the laser beam.

The 193-nm blocking filter can be removed so that the Rayleigh scattering is also imaged onto the camera. Figure 2(b) shows a single-laser-shot image with the wavelength tuned to the two-photon \( \text{H}_2 \) resonance. In this case the intensity is a combination of \( \text{H}_2-\text{O}_2 \) fluorescence and the UV Rayleigh scattering. Of particular interest are the thin, approximately vertical dark bands bordering the air portions of the flame. These bands define a sharp boundary of high temperature where the density, and therefore the Rayleigh scattering intensity, is reduced. As one proceeds closer to the combustion zone, the increasing temperature results in increased \( \text{O}_2 \) hot-band fluorescence.\(^5,6\) Figure 2(c) is the same as Figs. 2(a) and 2(b), except that the laser is now tuned away from both the \( \text{H}_2 \) and the \( \text{O}_2 \) resonances. In this case the image is dominated by Rayleigh scattering and therefore reflects the density.

Fig. 2. Instantaneous flame image illustrating the simultaneous imaging of two-photon LIF of \( \text{H}_2 \) and single-photon LIF of hot \( \text{O}_2 \). (b) Single-laser-shot image of flame under identical conditions as for (a), except that the 193-nm blocking filter is removed. The result is a combination of \( \text{H}_2, \text{O}_2 \) LIF, and Rayleigh scattering. (c) Single-laser-shot image of a flame as for (a) and (b), except that the ArF laser is tuned off both \( \text{H}_2 \) and \( \text{O}_2 \) resonance. The resulting intensity is dominated by Rayleigh scattering.

Figure 2(a) illustrates a typical single laser shot image from the \( \text{H}_2 \)-air diffusion flame, with the 7910 filter in place to block the Rayleigh scattering. The two-photon LIF from the central core of unburnt \( \text{H}_2 \) fuel can be clearly distinguished from the single-photon LIF associated with the hot air surrounding the jet. The dark region between the air and the fuel is the reaction zone where both air and fuel are rapidly consumed. Moving the laser waist toward the hot-air zone reduces the \( \text{H}_2 \) signal somewhat in the core, but the boundary between air and fuel remains distinct, indicating a lack of \( \text{H}_2 \) as opposed to merely a loss of signal resulting from the intensity drop-off of the pump beam. It also appears that there is a slight

![Figure 2](image)

![Figure 3](image)
profile weighted by the Rayleigh cross section. The H₂ fuel image is very weak (approximately a factor of 5 lower than that of air⁷) owing to its low Rayleigh scattering cross section.

We obtained excitation spectra in order to verify the previously cited spectroscopic assignments and to determine an approximate temperature of the air-rich region. Figure 3 shows two sets of experimental spectra along with O₂ LIF modeling predictions at 800 K. The spectra appear somewhat noisy owing to the high level of turbulence. The experimental spectra were obtained by spatially averaging the digitized signal from an approximately 10 × 10 pixel box centered in the air- and fuel-rich zones, respectively, of the flame. For the bottom spectrum, which is from the air region, the laser was scanned through 160 cm⁻¹ at a rate of approximately 23 cm⁻¹/min, resulting in a 12,600-frame image sequence. The major O₂ transitions are also labeled, and, except for the weakness of the P(21)/R(23) (7,1) transition pair at ∼193.29 nm, the agreement between the modeled and experimental O₂ spectra is quite good. The P(21)/R(23) (7,1) transitions are missing because of the close overlap with the O₂ P(17) (4,0) transition, which, because of strong absorption, causes the ArF laser output at this wavelength to be reduced and the locking efficiency to be poor. (N₂ purging of the laser and the beam path could be used if these lines are desired, as might be the case when accurate temperature measurements are required.) The top spectra, which are from both air and fuel regions, were obtained similarly but with a slower 25-cm⁻¹ scan in the region of 193.25 nm. It can be seen that the H₂ transition overlaps the high-wavelength shoulder of the R(17) (10,2) O transition at approximately the half-intensity point.

It is useful to discuss briefly the sensitivity limits and issues associated with quantifying the two-photon H₂ LIF imaging. From Ref. 8, the steady-state population of H₂ in the E, F state is given by

\[
[H₂^*] = \frac{α^2}{h_0 + σ_{pl}} [H₂], \tag{1}
\]

where \(α = \frac{σ}{I}\) is the two-photon absorption coefficient, \(σ_{pl}\) is the cross section for photoionization from the E, F state, and \(τ_{eff} = 1.8\) ns is the effective lifetime of the E, F state. This expression, as discussed in Ref. 8, assumes that the dominant collisional quenching channel for the E, F state is to the nearly degenerate (ΔE = 22 cm⁻¹) C¹Π₂ state, which then radiates to the X state.

The minimum detectable H₂ concentration can be estimated from Eq. (1) by using literature values for the constants and for the E, F-state radiative lifetime.⁸ We arrive at approximately 10² photoelectrons per pixel for H₂ at STP, assuming that a 50-mJ pulse, 1 cm × 0.01 cm sheet is 1:1 imaged onto a 250 × 250 array (40 µm × 40 µm pixel size), using f/2 collection optics and a 0.10 quantum efficiency. Assuming a conservative camera sensitivity of 1000 photoelectrons per pixel, this indicates that densities of the order of 0.01 amagat should be readily detectable.

While the potential for H₂ imaging is clearly promising, issues such as species-dependent quenching, temperature-density ambiguity, and laser intensity dependence must be resolved if quantitative measurements are to be performed. On the positive side, since H₂ is a major species, the measurement should not be significantly affected by laser-induced photochemical effects, which alter the species concentration.

While it is not the subject of this Letter, calculations by Huo¹⁰ show that the E, F ← X two-photon cross section for \(υ^* = 1\) is a factor of 2-3 larger than that for \(υ^* = 0\). This is significant in that it suggests that the B ← E, F fluorescence should be intense enough to use for the interrogation step of a H₂ RELIEF velocimetry measurement, similar to the O₂ RELIEF method described by Miles et al.¹¹

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