Two-dimensional imaging of molecular hydrogen in H_2–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 H_2–air diffusion flame. Excitation spectra of fuel and oxidant-rich flame zones confirm a partial overlap of the two-photon H_2 (6,0) $Q(1)$ $E, F \leftarrow X$ and single-photon O_2 (10,2) $R(17)$ $B \leftarrow X$ (Schumann–Runge) absorption bands. UV Rayleigh scattering images of flame structure and estimated detection limits for the H_2 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 H_2–O_2 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 H_2 excitation wavelength with a strong hot-band absorption allows for simultaneous imaging of high-temperature O_2 by using a single laser frequency. This is potentially significant for studies of mixing in turbulent H_2–air combustion, such as in supersonic combustion ramjet engines.

The H_2 excitation/fluorescence scheme, illustrated in Fig. 1, is that first described by Kligler and Rhodes.[1] The two-photon excitation is from the $v'' = 0$, $J'' = 1$ level of the $X\ ^1\Sigma_g^+$ state to the $v' = 6$, $J' = 1$ level of the $E, F\ ^1\Sigma_g^+$ state. The two-photon energy is 103 487 cm^{-1},[2] 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,[3] who conclude, based on the number of nodes in the vibrational wave function, that the level labeled $v' = 2$ by Kligler and Rhodes is more correctly labeled $v' = 6$.

The observed fluorescence is from the $v' = 6\ E, F$ $^1\Sigma_g^+$ state to the $v'' = 1$ and $v'' = 0$ levels of the $B\ ^1\Sigma_u^+$ state at 830 and 750 nm, respectively. The O_2 hot-band absorption is from the (10,2) $R(17)$ $B \leftarrow X$ 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 H_2 fluorescence by simple colored-glass filters. This allows instantaneous, separate H_2 and O_2 images to be obtained by using either split-screen techniques[4] 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 H_2–air diffusion burner constructed by flowing H_2 through a 1.5-mm

i.d. stainless-steel tube. The backing pressure is approximately 2000 Torr, and the flow rate is approximately 18 L/min. Fluorescence is collected normal to the laser propagation direction with an $f/6$ solid angle and imaged onto a dual-microchannel plate intensified charge-injection-device video camera that captures images at the standard 30-Hz video framing rate. A Corning 7910 glass filter is used to separate the LIF from the Rayleigh scattering. Alternatively, the filter can be removed, and, with the laser tuned off resonance, Rayleigh scattering can be made to dominate. The intensifier was designed for UV imaging, so its quantum efficiency is only 3% at 750 nm and less than 1% at 830 nm. Nonetheless, this efficiency was sufficient for instantaneous images of the turbulent H_2 flow to be obtained.

The images are recorded on video tape and subsequently digitized with a frame grabber (Data Translation Model DT-1451) controlled by a SUN 4/330 workstation. Excitation spectra are obtained by tuning the ArF laser oscillator grating with a stepper-motor-driven gear assembly. Digitization of specific portions of the resulting image sequence yields excitation spectra of different regions of the flame. The wavelength axis was determined by comparing the experimental O_2 transitions with known spectroscopic constants.
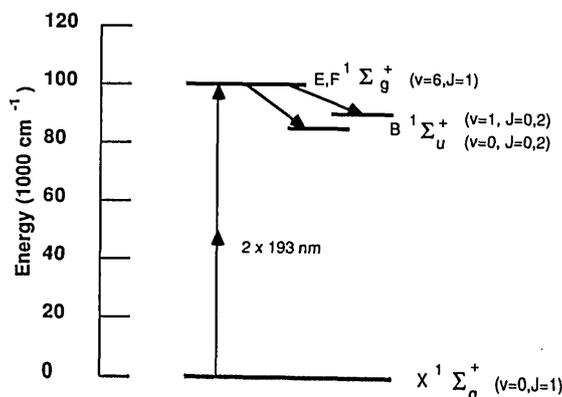

Fig. 1. H_2 energy-level diagrams for the relevant states for two-photon LIF.
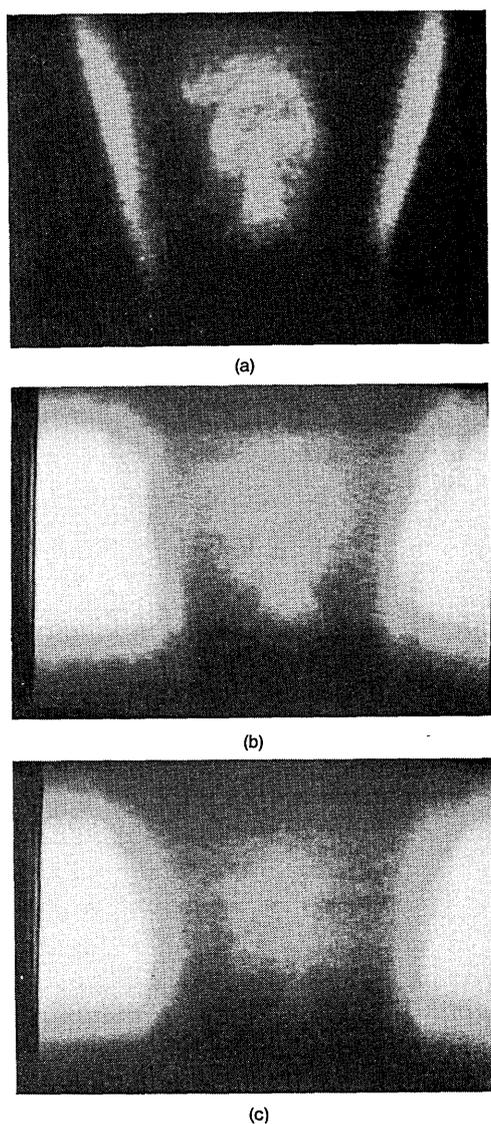
(a)

(b)

(c)

Fig. 2. Instantaneous flame image illustrating the simultaneous imaging of two-photon LIF of H_2 and single-photon LIF of hot 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 H_2, 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 H_2 and O_2 resonance. The resulting intensity is dominated by Rayleigh scattering.

Figure 2(a) illustrates a typical single laser shot image from the 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 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 H_2 signal somewhat in the core, but the boundary between air and fuel remains distinct, indicating a lack of 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

asymmetry in the 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 H_2 resonance. In this case the intensity is a combination of H_2–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 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 H_2 and the O_2 resonances. In this case the image is dominated by Rayleigh scattering and therefore reflects the density
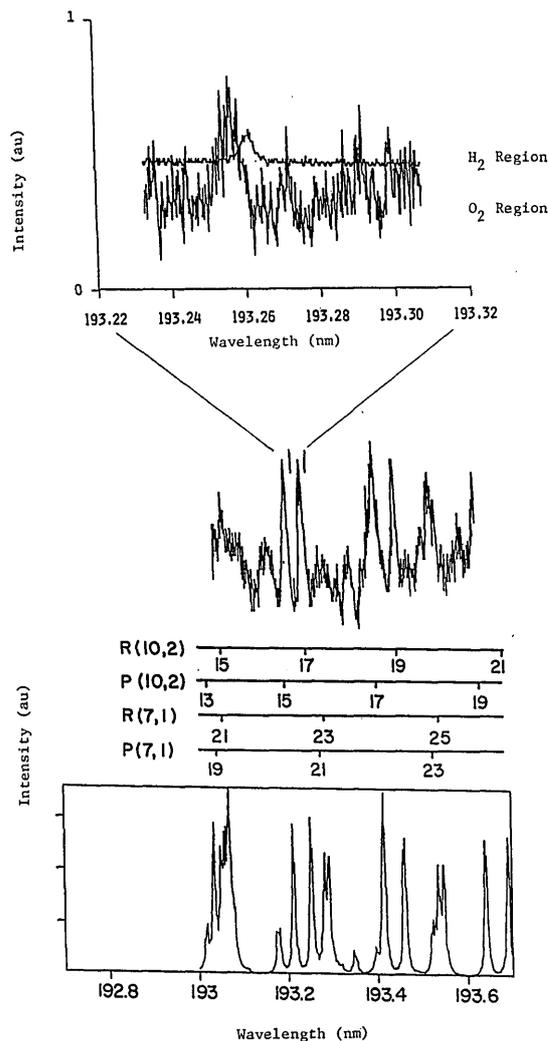

Fig. 3. Experimental (middle trace) O_2 excitation spectrum along with the 800-K modeling (lower trace) prediction and expansion (upper trace) of the 193.26-nm region illustrating the H_2–O_2 overlap.

profile weighted by the Rayleigh cross section. The H_2 fuel image is very weak (approximately a factor of 5 lower than that of air[7]) 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 O_2 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 O_2 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 O_2 spectra is quite good. The $P(21)/R(23)$ (7,1) transitions are missing because of the close overlap with the O_2 $P(17)$ (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. (N_2 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 H_2 transition overlaps the high-wavelength shoulder of the $R(17)$ (10,2) O_2 transition at approximately the half-intensity point.

It is useful to discuss briefly the sensitivity limits and issues associated with quantifying the two-photon H_2 LIF imaging. From Ref. 8, the steady-state population of H_2 in the E, F state is given by

$$[\text{H}_2{}^*] = \frac{\alpha I^2}{\dfrac{\hbar\omega}{\tau_{\text{eff}}} + \sigma_{\text{pi}}I} [\text{H}_2], \qquad (1)$$

where $\alpha = \sigma/I$ is the two-photon absorption coefficient, σ_{pi} is the cross section for photoionization from the E, F state, and $\tau_{\text{eff}} = 1.8$ 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 ($\Delta E = 22$ cm^{-1}) $C^1\pi_u$ state, which then radiates to the X state.

The minimum detectable H_2 concentration can be estimated from Eq. (1) by using literature values for the constants[2,3,9] and for the E, F-state radiative lifetime.[8] We arrive at approximately 10^5 photoelectrons per pixel for H_2 at STP, assuming that a 50-mJ pulse, 1 cm $\times$ 0.01 cm sheet is 1:1 imaged onto a 250×250 array (40 μm $\times$ 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 H_2 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 H_2 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 Huo[10] show that the $E, F \leftarrow X$ two-photon cross section for $v'' = 1$ is a factor of 2–3 larger than that for $v'' = 0$. This is significant in that it suggests that the $B \leftarrow E, F$ fluorescence should be intense enough to use for the interrogation step of a H_2 RELIEF velocimetry measurement, similar to the O_2 RELIEF method described by Miles et al.[11]

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