Appl Phys B (2009) 96: 241–245
DOI 10.1007/s00340-009-3597-6

Applied Physics B
Lasers and Optics

RAPID COMMUNICATION

Hydroxyl radical imaging at kHz rates using a frequency-quadrupled Nd:YLF laser

M.E. Cundy · V. Sick

Received: 31 March 2009 / Revised version: 30 April 2009 / Published online: 11 June 2009

Abstract Laser-induced fluorescence imaging of hydroxyl radicals has been an important tool in combustion research for more than twenty years. More recently, high frame rate imaging of hydroxyl radicals has been demonstrated using Nd:YAG-pumped dye laser systems. This work describes how a high repetition rate frequency-quadrupled Nd:YLF laser emitting at 263 nm can be used for laser-induced fluorescence imaging of hydroxyl radicals with less complexity. Hydroxyl radicals are excited in the A–X (2,0) band and redshifted fluorescence emission is detected with an image intensified CMOS camera at kHz frame rates. Furthermore, a strategy for high-speed temperature imaging is described.

PACS 42.62.Fi · 33.50.Dq · 82.33.Vx

1 Introduction

Hydroxyl radicals have been and continue to be the target of numerous investigations for their role in atmospheric chemistry [1] and combustion [2]. Their high reactivity makes them important to many reactions in the atmosphere and in chain branching reactions in flames [2]. Amongst a range of suitable techniques, laser-induced fluorescence (LIF) is the most commonly used to detect OH in flames [3]. This is in part due to the ability to capture images of OH distributions. The diatomic OH radical has a pronounced well-resolved rotational–vibrational spectrum that has been characterized comprehensively [4] and allows quantum-state specific excitation. LIF detection requires excitation with a laser source that will excite OH to the electronic A state. Typical transitions that have been explored occur at wavelengths between 248 and 320 nm. Tunable dye laser excitation provides the widest flexibility to choose a specific excitation wavelength. Numerous studies have also been performed using tunable excimer lasers. Their simplicity and high pulse energy allowed applications with excitation using weaker transitions, such as the (3,0) band (KrF excimer at 248 nm) and the (0,0) band (XeCl excimer at 308 nm) [5, 6]. More recently, a frequency-quadrupled Nd:YAG laser with an intracavity etalon was used to excite OH in the (2,0) band [7]. This study also showed that fuel LIF tracers, such as acetone or 3-pentanone, can be simultaneously detected with OH radicals. A similar approach was demonstrated with a Nd:YAG pumped dye laser system [8] to study the equivalence ratio near the flame front. Such simultaneous OH/fuel measurements will be important to investigate ignition stability and local combustion progress as a function of mixture conditions in reciprocating engines. In light of the stochastic nature of turbulent mixing and combustion processes, measurements that resolve relevant time and length scales are important. This means that at least two-dimensional information should be obtained at kHz frame rates. Fuel imaging using LIF has been demonstrated at 12 kHz based on biacetyl as a fuel tracer and excitation with a frequency-tripled diode-pumped Nd:YAG laser [9]. OH LIF imaging was demonstrated at a rate of 5 kHz using an Nd:YLF laser pumped dye laser to study turbulent flames [10], and the same diagnostics were later used in combination with particle image velocimetry [11]. A burst-mode optical parametric oscillator was used to excite OH radicals at 313.5 nm at up to 50 kHz, but sequences of only up to 100 images can presently be obtained [12]. The 283 or 313.5 nm light used

M.E. Cundy (✉) · V. Sick
Department of Mechanical Engineering, 2026 W.E. Lay Automotive Laboratory, The University of Michigan, 1231 Beal Ave, Ann Arbor, MI 48109-2133, USA
e-mail: mcundy@umich.edu
Fax: +1-734-764-4256

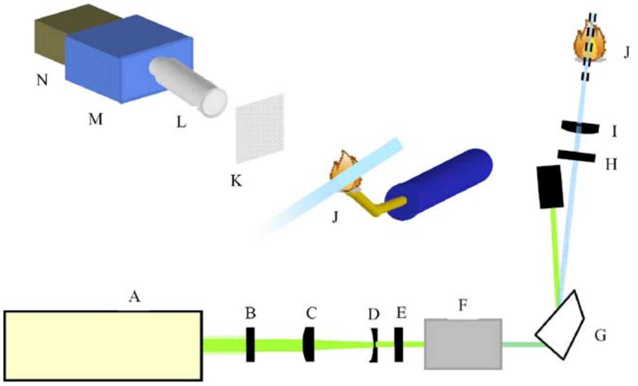

Fig. 1 Schematic of the experimental setup. The camera/filter/flame setup is shown as an enlarged insert. Explanations of the symbols can be found in the text

for OH excitation could also be used simultaneously for fuel tracer LIF measurements as well. However, a simplification of instrumentation for simultaneous OH/fuel measurements is desirable.

An inspection of the OH A–X (2,0) band indicates that several absorption lines should fall within the bandwidth of a frequency-quadrupled Nd:YLF laser. This paper presents a feasibility study on the applicability of a frequency-quadrupled Nd:YLF laser operating without an intracavity tuning element to produce LIF signals of OH radicals. A demonstration of the technique is given with images obtained in a propane torch flame.

2 Experimental details

The experimental setup, shown in Fig. 1, includes a dual-cavity, frequency-doubled, diode-pumped Nd:YLF laser (Quantronix Darwin Duo; A). The two beams exiting the laser head are circularly polarized in opposing directions. A zero order achromatic quarter waveplate (Newport; E) was used to produce linearly polarized light, where the polarizations of the two beams are orthogonal to each other. Therefore, only one of the lasers was used for frequency-doubling from green to UV and the second laser was not triggered during the experiments. The green beam, at a power of 24 W was passed through an aperture (B) and focused with a Galilean telescope [plano-convex lens (C), $f = +75$ mm; plano-concave lens (D), $f = -25$ mm] to a beam diameter of ~3 mm. It was then sent through a KD*P crystal (Quanta-Ray HG-2; F) taken from a pulsed Nd:YAG laser system (Spectra Physics DCR-11). Green and UV beams were separated with a Pellin–Broca prism (G), and the UV beam is passed through a cylindrical lens ($f = 75$ mm; H) and plano-convex focusing lens ($f = +500$ mm; I) to produce a light sheet ~7 mm tall and 0.5 mm wide at the flame location (J).

A propane soldering torch was used as a simple and convenient source of OH radicals in a flame.

The laser was operated at 1 kHz in this work to demonstrate that OH can be excited at 263 nm. Green pulse energies of 24 mJ generated UV pulse energies of 50 µJ. Laser-induced fluorescence emission after excitation within the A–X (2,0) band is expected from 260 to ~330 nm with the strongest emissions occurring between 280 and 330 nm [7]. OH LIF signals are by far the dominating signals in this wavelength range in a gas flame and simple broadband filtering can be used for detection. A spectral range for the LIF signals was identified by sequentially using color glass filters (Schott WG 280 and WG 365; K) in front of the imaging lens (Coastal Optics; L) on the intensifier (HS-IRO, LaVision; M), which was lens-coupled to a high-speed CMOS camera (Vision Research Phantom 7.3; N). Filtering with a WG 365 filter will block any OH fluorescence and therefore will allow to positively identify OH LIF signal. Furthermore, due to the low pulse energy, high intensifier gains have to be used and the contamination of the LIF images with chemiluminescence is likely. The difference of images taken sequentially with a WG 280 and WG 365 filter placed in front of the intensifier leaves net images of OH LIF signals.

3 Results and analysis

The image sequence shown in Fig. 2 demonstrates that selective LIF detection of OH was achieved with a frequency-quadrupled Nd:YLF laser. Figure 2(a) shows signals filtered with a Schott WG 280 color glass filter. The image shows two oval areas on the periphery of the jet flame at the location where the light sheet intersects the flame horizontally. Due to the low pulse energy that was available the image intensifier was set to a high gain of 9,000. Thus, despite the short gate time of 400 ns the image contains some chemiluminescence signals at longer wavelengths, such as from CH^* and maybe to a lesser extent C_2^* in the core of the flame where the inner premixed flame is located. With the same intensifier settings, a 200-image average of signals was measured with a Schott WG 365 color glass filter installed. This image is shown in Fig. 2b. The OH LIF signals are absent but the longer wavelength chemiluminescence signals are still present. The difference of the images from Figs. 2a and b is displayed in Fig. 2c and clearly shows the successful isolated detection of OH LIF signals at the expected location in the flame.

The averaged images shown in Fig. 2 were obtained using excitation energies of 50 µJ per pulse. This is the same pulse energy that Kittler and Dreizler [10] reported for their OH LIF work using a high repetition rate dye laser system operating at 283 nm. Based on the Einstein B coefficients for absorption the expected signal ratio between excitation

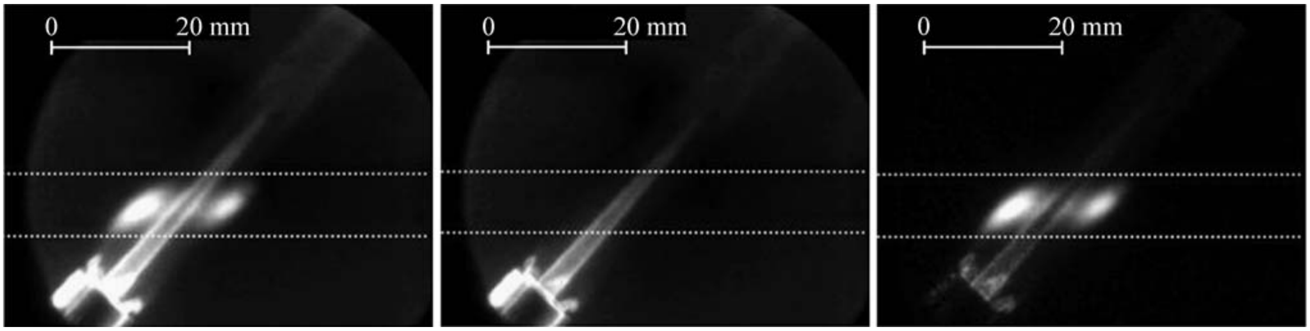

Fig. 2 Average of 200 images taken with the WG 280 filter (*left*) and WG 365 filter (*center*). The difference of the two images (*right*) shows largely OH LIF signals to the left and right of the inner core of the flame along with some residual chemiluminescence signal. Note that the intensifier settings were 400 ns gate time and a gain of 9,000 to obtain OH LIF signals with only 50 µJ of broadband light at 263 nm. The extent of the light sheet is marked with the *dotted white lines*

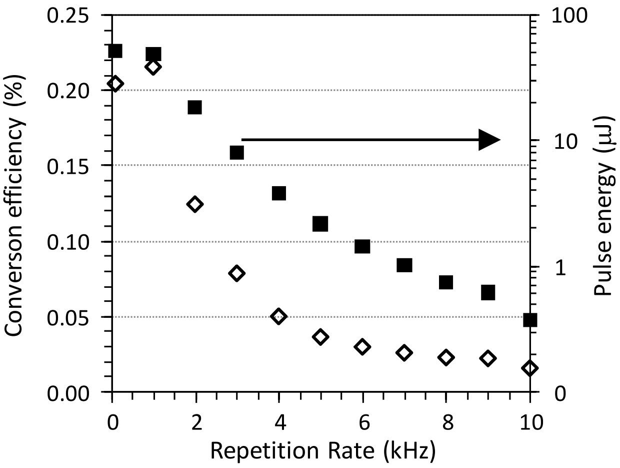

Fig. 3 Conversion efficiency (◇) and pulse energy (■) at 263 nm rapidly decrease with increasing repetition rate (and accordingly pulse width)

in the (1,0) band at 283 nm and the (2,0) band at 263 nm is about a factor of 6 in favor of the (1,0) excitation [13]. Increasing the pulse energy of the quadrupled Nd:YLF to just a mere 300 µJ will enable measurements of similar quality as reported with the dye laser system. The pulse energy of the system that was used in this work depends on the repetition rate and is shown in Fig. 3. The pulse energy and the conversion efficiency from 527 to 263 nm decrease with increasing repetition rate. Overall, the conversion efficiency is low for the present system. However, this is not unexpected given that the Nd:YLF emission is spectrally quite broad ($\sim$20 cm^{-1} at 527 nm) and the pulse duration of the 527 nm light increases from 100 to 400 ns at 1 and 10 kHz, respectively. The intensity of the frequency-doubled light for low conversion rates is given as [14]:

$$I(2\omega, L) = \frac{I^2(\omega)(2\omega^2|\chi_{\text{eff}}^{(2)}|^2 L^2)}{n^3 c^3 \varepsilon_0} \frac{\sin^2(\Delta k L)}{(\Delta k L)^2} \tag{1}$$

where I is the intensity, ω is the frequency of the fundamental wave, L is the shorter of the coherence length or length of crystal, $\chi_{\text{eff}}^{(2)}$ is the effective second-order nonlinear susceptibility tensor, n is the index of refraction of the crystal material, c is the speed of light, ε_0 is the permittivity of free space, and Δk is the difference between the sum of the two ordinary wave vectors and the extraordinary wave vector which is related to the phase difference. The conversion efficiency is therefore expected to increase if the focus and second-order nonlinear susceptibility tensor are increased and if pulse duration and bandwidth are reduced. The former two can be accomplished using different lenses and a BBO or CLBO crystal, and it is estimated that 300 µJ pulses at 1 kHz can be achieved. Clearly, if all options are pursued, much higher pulse energies can be expected and single-shot imaging at high frame rates and high signal-to-noise ratio will be enabled.

The origin of the OH LIF signals is due to excitation of several rotational transitions within the A–X (2,0) band. The fundamental spectral bandwidth of a Nd:YLF is around 360 GHz [15] and thus at the fourth harmonic laser emission can be expected from around 263.1 to 263.4 nm. A LIFBASE [13] simulation of an excitation LIF spectrum in this range is shown in Fig. 4 for atmospheric pressure conditions at 2,000 K. A clear rotational structure is visible, including transitions from rotational levels ranging from $N'' = 3$ to 20. A more in-depth analysis will have to be performed in the future to determine the exact overlap of the frequency-quadrupled emission of the Nd:YLF laser and the OH spectrum. To this extent, the use of an intracavity etalon in the Nd:YLF laser can be useful to tune the laser across the absorption lines. This will help to increase signal and will enable temperature imaging measurements as described below. This analysis will also be needed to determine temperature dependence of the LIF signals. Details of this and a discussion of collisional effects on OH LIF from $v' = 2$ can be found in [7].

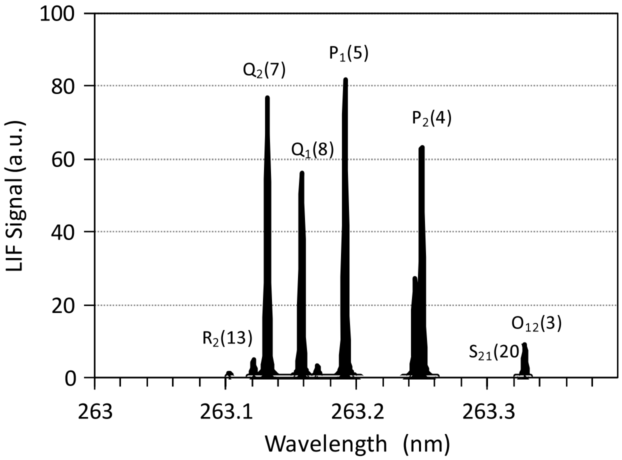

Fig. 4 Simulated OH LIF spectrum that lies within the range of the fourth harmonic of the 1,053 nm Nd:YLF emission. The simulation was carried out with LIFBASE v2. For clarity, only selected rotational lines are labeled. The bandwidth of the fourth harmonic is expected to approximately cover the displayed wavelength range

The overlap of the frequency-quadrupled Nd:YLF laser output with a number of OH rotational lines opens opportunities to devise a convenient tool to measure temperature distributions using LIF of OH radicals. Two-line single-shot OH LIF measurements have been described in the literature [16–18], but all measurements were conducted at a repetition rate of a few Hz. On the other hand, high repetition rate OH LIF measurements were demonstrated but not for temperature measurements [10]. For instantaneous temperature imaging two Nd:YLF/dye laser systems would be required. An alternative approach could be taken using intracavity line narrowing of an Nd:YLF laser, e.g., using an etalon, to enable tunability and specific excitation from a selected rotational level. In a dual-cavity laser system, such as the one used in this study, both resonators could be equipped with etalons allowing independent tuning of each laser. The overlapped frequency-doubled beams are cross-polarized and therefore each can be individually frequency-doubled to 263 nm. The combination of low and high rotational quantum number excitation results in good temperature sensitivity and thus accuracy of the temperature measurement [16]. It has to be pointed out, however, that high-speed CMOS cameras need to be carefully calibrated to ensure that the ratio of two images taken with two separate cameras is not biased by offsets, different nonlinearities, and sensitivities for each pixel. Progress in camera technology can be expected to facilitate this task.

4 Conclusions

High-speed imaging techniques are becoming increasingly important for investigations of combustion and fluid mechanics problems. Resolving relevant temporal and spatial scales requires imaging at kHz rates. Camera technology is available to achieve these rates but there are still limitations in laser-based excitation sources, especially when tunability is required for specific excitation of select molecules. High repetition rate dye lasers are available and have been used for example to demonstrate LIF detection of hydroxyl (OH) radicals [10]. In the present work, the use of a frequency-quadrupled Nd:YLF laser, operating at 263 nm, for selective excitation of OH radicals at kHz rates was demonstrated. OH LIF images were acquired from a flame of a propane torch at 1 kHz frame rate with 50 µJ per pulse. It was shown that a group of rotational OH lines can be excited with an unmodified frequency-quadrupled Nd:YLF laser. The implementation of an intracavity tuning element into the Nd:YLF laser seems advantageous to add tunability and to potentially increase the pulse energy of the frequency-quadrupled light. It is also expected that shorter pulse duration would help to increase the conversion efficiency of fourth-harmonic generation. The prospect of increased selectivity for excitation of individual rotational transitions was discussed and the potential for single-shot high-speed temperature imaging using a dual-cavity laser was presented.

Acknowledgements This work was supported through the General Motors Collaborative Research Laboratory on Engine Systems Research at the University of Michigan and a graduate student research fellowship from the University of Michigan.

References

1. J. Barker (ed.), *Progress and Problems in Atmospheric Chemistry*, vol. 3 (World Scientific, Singapore, 1995)
2. J. Warnatz, U. Maas, R.W. Dibble, *Combustion*, 2nd edn. (Springer, Berlin, 1999)
3. K. Smyth, in *Applied Combustion Diagnostics*, ed. by K. Kohse-Höinghaus, J.B. Jeffries (Taylor and Francis, New York, 2002)
4. G.H. Dieke, H.M. Crosswhite, J. Quant. Spectrosc. Radiat. Transf. **2**, 97–200 (1962)
5. W. Ketterle, M. Schäfer, A. Arnold, J. Wolfrum, Appl. Phys. B **54**, 109–112 (1992)
6. E.W. Rothe, P. Andresen, Appl. Opt. **36**, 3971–4033 (1997)
7. V. Sick, N. Wermuth, Appl. Phys. B **79**, 139–143 (2004)
8. D.A. Rothamer, J.B. Ghandhi, SAE Paper 2003-01-0069 (2003)
9. J.D. Smith, V. Sick, Appl. Phys. B **81**, 579–584 (2005)
10. C. Kittler, A. Dreizler, Appl. Phys. B **89**, 163–166 (2007)
11. B. Böhm, C. Heeger, I. Boxx, W. Meier, A. Dreizler, Proc. Combust. Inst. **32**, 1647–1654 (2009)
12. J.D. Miller, M. Slipchenko, T.R. Meyer, N. Jiang, W.R. Lempert, J.R. Gord, Opt. Lett. **34**, 1309–1311 (2009)
13. J. Luque, D.R. Crosley, LIFBASE v2.0.2 ed. (SRI International, MP-99-0099, http://www.sri.com/psd/lifbase 1999)
14. W. Demtröder, *Laser Spectroscopy: Basic Concepts and Instrumentation*, 2nd edn. (Springer, Heidelberg, 1998)
15. G.P.A. Malcolm, P.F. Curley, A.I. Ferguson, Opt. Lett. **15**, 1303–1305 (1990)

16. J.M. Seitzmann, R.K. Hanson, Appl. Phys. B **57**, 385–391 (1993)
17. U.E. Meier, D. Wolff-Gaßmann, W. Stricker, Aerosp. Sci. Technol. **4**, 403–414 (2000)
18. A. Arnold, B. Lange, T. Bouché, Z. Heitzmann, G. Schiff, W. Ketterle, P. Monkhouse, J. Wolfrum, Ber. Bunsenges. Phys. Chem. **96**, 1388–1392 (1992)