TECHNICAL NOTE

Tunable KrCl excimer-laser operation for combustion diagnostics

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Operating conditions for a tunable XeCl excimer laser have been optimized for tunable narrow-band operation at 222 nm with KrCl, formed in Kr/He/Ne/HCl gas mixtures. This wavelength is interesting for detection of nitric oxide (NO) in combustion environments. The laser emission coincides with the (1, 1) and (2, 2) bands of the NO *A–X* system. Laser-induced-fluorescence spectra taken in a flame show specific excitation of NO with this laser.

Key words: Excimer laser, combustion diagnostics, laser-induced fluorescence.

Nitric oxide (NO) was identified as one of the most problematic pollutants formed in most combustion processes, and in recent years major progress has been made in understanding the kinetics of NO formation. However, there are still discrepancies between predictions made by mathematical models that use detailed treatment of chemical and fluid-mechanic properties of the combustion process and experimental data. Among other techniques, laser-based techniques have been especially helpful in giving new insight into NO formation in flames. Laser-induced fluorescence can also be used to measure two-dimensional concentration distributions, which is very valuable for experiments in turbulent combustion processes. For this technique high pulse energies are usually required. This is one of the reasons why tunable excimer lasers have become popular laser-light sources in this area. For a variety of molecules, such as OH, various aldehydes, NO_2, O_2, H_2O, and NO, the tuning range of different excimer lasers overlaps with suitable absorption bands. For NO, mainly ArF excimer lasers were used at 193 nm to excite transitions in the *D–X* band.[1] However, the use of this short wavelength is limited to a few cases in which absorption by oxygen and hydrocarbon species does not interfere with the laser. In this Note we address the operation of a tunable KrCl excimer laser. We modified a tunable XeCl excimer laser to operate at 222 nm and show that we obtained narrow-band operation of this laser. Selective excitation of NO in the *A–X* band was achieved with this laser.

The excimer laser we used in our experiments was a Lambda Physik EMG 150 EST, without a magnetic switch control, that was running at a maximum repetition rate of 10 Hz. In a first series of experiments, we optimized the composition of the gas mixture for maximum energy output. Pulse energies were measured with an energy meter (Gentec ED 200). High-purity gases and gas mixtures were used. Kr, He, and Ne were of 99.9990% purity. HCl was supplied as a mixture of 5% HCl in He. First, only the oscillator resonator of the laser was used with a high-reflectivity mirror on one end and a mirror with 30% reflectivity on the output side. Second, the oscillator output was injected into the amplifier that was running with the same gas-mixture composition without any mirrors, thus yielding only single-pass amplification of the oscillator beam. The gas mixture of the amplifier was now optimized for maximum gain. Variation of all gases resulted in a bell-shaped curve for output energy versus partial pressure of the corresponding gas. Table 1 lists the most efficient gas mixtures as evaluated for the EMG 150 EST. It was found that using Ne as a buffer gas results in higher pulse energies compared with He. However, the output power when the Ne mixtures are used degraded within minutes to 38 mJ, whereas the He mixtures gained during time and remained at a constant pulse energy of 25 mJ for a longer time. These values are given for the broadband laser operation. We could

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Table 1. Mixture Composition for KrCl Operation of a EMG 150 EST Excimer Laser

Gas	Oscillator (mbars)	Amplifier (mbars)	Amplifier (mbars)
5% HCl/He	175	125	75
Kr	400	400	275
He	1800	1700	—
Ne	—	—	2350
Total Pressure	2375	2225	2700

not increase the peak pulse energy by setting the high voltage higher than 31 kV.

Tunability of the laser was achieved by removal of the high-reflectivity mirror and expansion of the oscillator beam with two or three MgF_2 prisms to illuminate a 600 lines/mm grating (Fig. 1). This is the standard grating delivered with the EMG series lasers.

When the oscillator was run with the optimized gas mixture for broadband operation, it was not possible to achieve stable lasing with all three prisms for expansion of the beam. We therefore removed one of the prisms at the cost of a higher bandwidth, because now only a fraction of the grating was illuminated. The oscillator beam was then injected into the amplifier resonator and amplified. The pulse energies obtained with this arrangement were approximately 1 mJ. To analyze the spectral width of the laser radiation, we focused a small fraction of the output beam onto the 100-μm entrance slit of a 1.3-m monochromator (McPherson 209, with a 2400 lines/mm grating). The monochromator grating was tuned to measure the spectral bandwidth. The signals were detected after the 200-μm exit slit with a photomultiplier tube (EMI 9783B) and integrated with a boxcar integrator.

Figure 2 compares the bandwidth of the broadband amplifier when equipped with the high-reflectivity mirror and the 30% outcoupling mirror with the narrow-band arrangement. The bandwidth decreases from 80 to 10 cm^{-1}. This is still too broad to excite single rotational lines of NO selectively at atmospheric pressure. But because NO has large broadening coefficients,[2] at higher pressures the rotational lines will blend, and thus resolution of single rotational lines is not possible anymore, even with a narrow-band laser. Still it will be advantageous to have a laser bandwidth narrower than the 80 cm^{-1} of the broadband KrCl excimer laser to minimize interference with the O_2 Schumann–Runge transitions.

To prove the applicability of this laser for laser-induced-fluorescence measurements of NO, we measured spectra excited in a small Bunsen burner flame. At 2000 K, the X state vibrational-level populations are approximately 1:0.25:0.065 for $v'' = 0$, 1, and 2, with the population of $v'' = 0$ set to 1. With comparable oscillator strengths for each of these bands,[3] the estimated sensitivity for the higher vibrational bands is still high enough to detect NO in unseeded flames. Because of the relatively poor locking efficiency of our laser setup (see Fig. 2 and

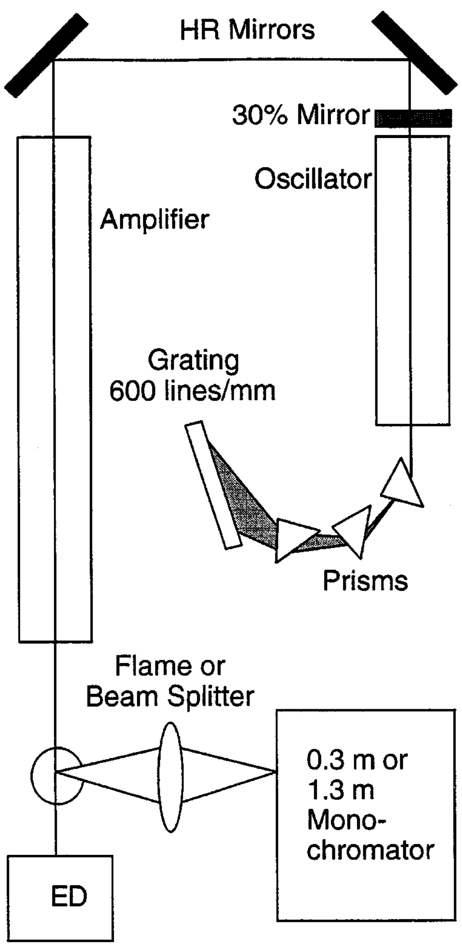

Fig. 1. Experimental setup for measurements of the laser bandwidth (beam splitter) and laser-induced fluorescence (flame). HR, high reflectivity; ED, energy detector.

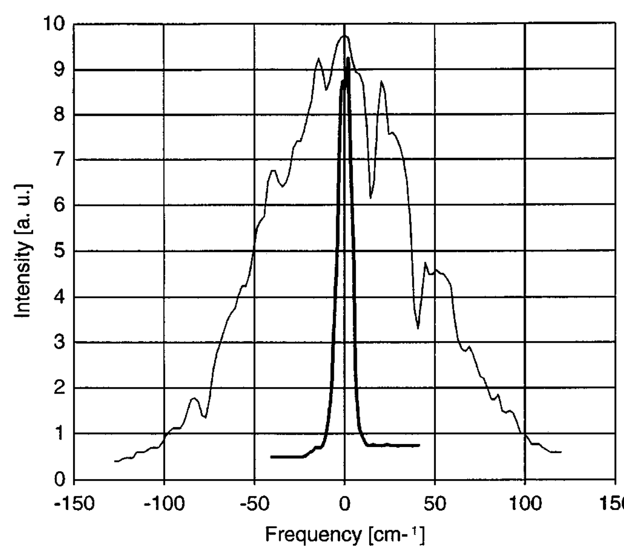

Fig. 2. Output intensity of the broadband KrCl excimer laser (thin curve) compared with the spectrally narrowed output (thick curve).

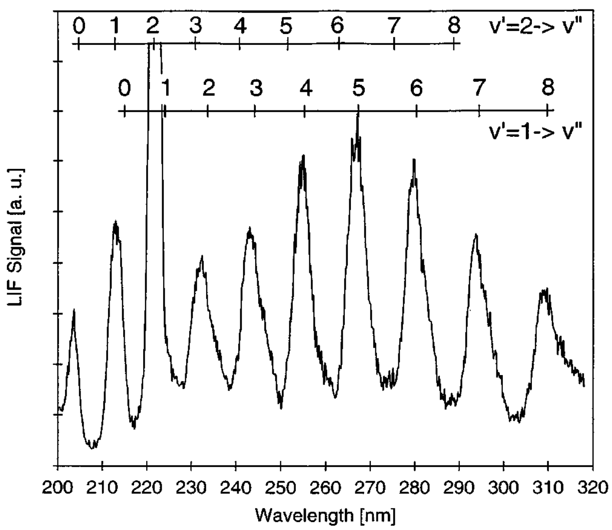

Fig. 3. Fluorescence spectrum of NO, excited with the narrow-band KrCl excimer laser. The two major progressions starting from $v' = 1$ and 2 are designated at the top. The peak at 222 nm is off the scale.

the text above), we were unable to see any rotational structure in excitation scans, even when the flame was seeded with NO. However, the emission spectrum plotted in Fig. 3 clearly shows that we selectively excited NO in this flame. The emissions can be assigned to the $(2, v'')$ and $(1, v'')$ progressions of the NO A–X system.

It has to be pointed out that the stability of the narrow-band laser output and the achievable pulse energies are projected to be substantially better when a newer tunable excimer laser is used. The reason for this is the short length of the oscillator (850 mm) of the laser that we used. This could be directly proved in our experiments when energy output of the oscillator was compared with the output of the amplifier when both were run in the broadband mode. Also, using the longer amplifier resonator of our laser as oscillator improved the stability and narrow-band output. Finally, we did not use any optics for the amplifier but operated it as a single-pass amplifier. The use of unstable resonator optics would increase the total energy output. With these improvements a tunable KrCl excimer laser will be a useful tool for combustion diagnostics. Because transitions of the (2, 2) and the (1, 1) bands are excited simultaneously, this excitation scheme can be used to measure temperatures with a single laser pulse by evaluation of the ratio of the fluorescence intensities emitted from $v' = 2$ and $v' = 1$. The Franck–Condon pattern of the two progressions[3] and the spectral separation of the vibrational bands permits the separation of the corresponding emissions from $v' = 2$ and $v' = 1$. This is obvious from Fig. 3, in which all the long-wavelength signals are due to fluorescence from $v' = 1$.

A tunable KrCl excimer laser provides a further excitation source for the detection of NO in addition to already available systems, such as ArF and KrF excimer lasers, in combination with Raman cells, tunable dye lasers, and, recently, optical parametric oscillators. Compared with excitation at 193 nm, interference by other absorbing species are reduced at 222 nm. The lower sensitivity compared with dye-laser pumping in the A–X (0, 0) band can be compensated by the higher pulse energies only at high temperatures, where the population of the ground state is high enough. For many technical applications, however, the qualitative detection of NO in high-temperature regions provides enough information for design purposes of combustion devices. Additionally, the simultaneously obtained temperature information can be used to correct for the temperature dependence. A strong argument for using tunable excimer lasers in general is the ease of use compared with Nd:YAG or excimer-laser-pumped dye lasers with subsequent frequency conversion, especially if transportability is one of the requirements.

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