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Development of a Compact, Narrow-linewidth, Tunable Ultraviolet Laser Source for Detection of Hg^0

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Abstract: A portable laser for real-time, stand-off detection of Hg^0 emissions from coal-fired power plants is developed and characterized. The pulse energy of the 254-nm laser is 1.8 μJ, which will enable sub-ppb detection of Hg^0.

OCIS codes: (190.2620) Harmonic generation and mixing; (280.1120) Air pollution monitoring; (140.3610) Lasers, ultraviolet

1. Introduction

Recent federal regulations targeting mercury emissions from coal-fired power plants have prompted increased activity towards the development of reliable, sensitive chemical sensors for monitoring mercury emissions. Current continuous mercury emissions monitors use extractive techniques requiring a probe and sampling lines to transfer the gas from the stack to the point of analysis. Multiple problems associated with the probe and sampling lines, such as plugging of the probe and chemical reactions within the sampling lines, degrade the measurement capabilities of these methods. In contrast, optical techniques are unique in their ability to analyze the flue gas *in situ*, thereby eliminating the challenges associated with sample extraction and conditioning. To meet the need for a real-time, non-invasive method for monitoring mercury emissions, we are developing laser-based techniques for short-range, sensitive detection of the two primary forms of vapor-phase mercury emitted by utility coal boilers, elemental mercury, Hg^0, and mercuric chloride, $HgCl_2$. The current paper examines the design and performance of a laser source and detection approach for Hg^0.

To monitor the concentration of Hg^0, we excite the Hg ($6\ ^3P_1 \leftarrow 6\ ^1S_0$) transition at 253.7 nm and detect the resulting resonant emission. In order to differentiate between the overlapping laser scatter and Hg^0 emission, an isotopically-pure Hg-202 atomic resonance filter (ARF) is used. The ARF absorbs spectrally-narrow light resonant with the laser frequency, while transmitting a portion of the broader Hg^0 emission. Consequently, the laser spectrum must be centered at and narrower than the Hg-202 ARF absorption feature at 253.7 nm. Performance modeling of this Hg^0 detection strategy indicates that the laser should have a linewidth ≤5 GHz (full width at half-maximum, FWHM) and a pulse energy ≥0.1 μJ. In addition, the laser must have the requisite physical characteristics (size, weight, power consumption) for use in a coal-fired power plant.

The current paper discusses a laser system designed to satisfy the aforementioned criteria based on the frequency conversion of the output of an optical parametric amplifier (OPA). The measured performance of the laser is compared to theoretical simulations, and the performance of the ARF with the laser is evaluated.

2. Experimental

The laser architecture chosen for the Hg^0 sensor involves multiple stages of nonlinear frequency conversion, as depicted in Fig. 1. To select the optimum nonlinear crystals and focusing conditions, we simulate the nonlinear mixing processes using SNLO [1], a freely available software package developed at Sandia National Laboratories. SNLO calculates the expected output pulse energy, as well as spatial, temporal, and spectral profiles. The description of the system designed using the results of these calculations follows.

The fundamental output of a passively Q-switched, monolithic Nd:YAG microlaser [2], pumped by a 45-W fiber-coupled diode laser (Jenoptik, JOLD-45-CPXF-1L) and operating at 10 Hz, is frequency doubled in LBO (Castech, 10-mm long, XY cut, θ = 90°, ϕ = 12°) using Type I phase matching. The resulting 532-nm beam is used to pump the OPA. The OPA is MgO-doped, periodically-poled $LiTaO_3$ (HC Photonics Corp., 10-mm long, period: 9.11 μm) temperature stabilized to ~40°C. $LiTaO_3$ is chosen over periodically-poled $LiNbO_3$ for its higher damage threshold and lower operating temperature. The OPA is seeded by a fiber-coupled, tunable, cw distributed feedback diode laser (Sacher Lasertechnik Group, SYS 050-DFB-0760.98-40) tuned to 761 nm. The seed laser has a tuning range of 2 nm. The amplified 761-nm signal beam is Type I frequency doubled in two LBO crystals (Castech, 10-mm long, XY cut, θ = 90°, ϕ = 36.5°) in a walkoff-compensating geometry. Finally, 254-nm light is generated

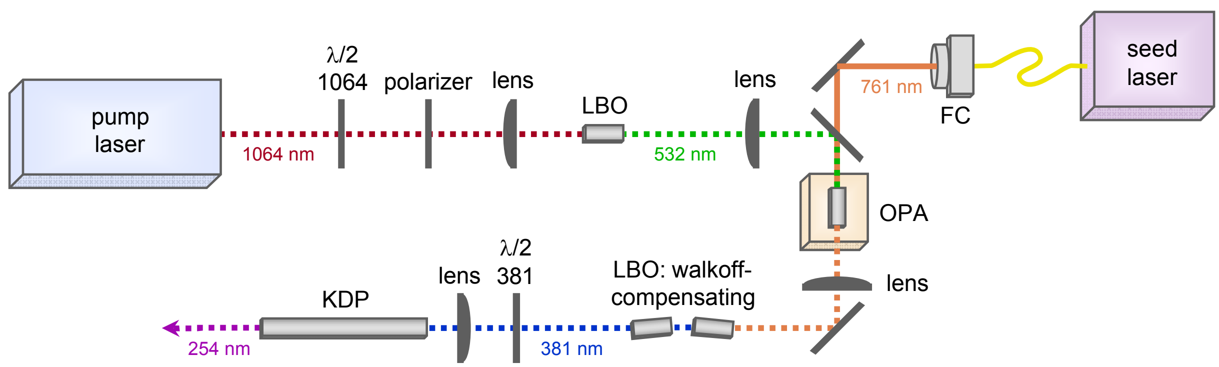

Fig. 1. Diagram of laser for Hg^0 sensor. FC, fiber coupler; λ/2, half-wave plate (at the designated wavelength in nm). Straight dashed and solid lines denote free-space pulsed and cw beams, respectively. The curved yellow line represents a fiber. The laser system is mounted on a 45.7 cm × 30.5 cm breadboard.

by Type I mixing of the 761-nm and 381-nm beams in KDP (Castech, 50-mm long, 76.5°). A zero-order waveplate (full-wave at 761 nm, half-wave at 381 nm) is placed between the walkoff-compensating LBO crystals and the KDP crystal to rotate the polarization of the 381-nm beam to be parallel to that of the 761-nm beam. The nonlinear crystals are positioned near the beam waists provided by the lenses and are angle tuned to maximize the 254-nm pulse energy. The frequency conversion efficiencies, spectral properties, and spatial profiles of the laser are characterized using pyroelectric energy meters, an optical spectrum analyzer (OSA), and a CCD camera.

The isotopically pure Hg^0 ARF (Opthos Instruments, Inc.) used to filter the narrow linewidth scatter contains 10 mg of Hg-202 with an isotopic purity of 96% in a bath gas of 76 torr O_2. The O_2 is added to spectrally broaden the absorption feature and to effectively quench emission from the Hg-202 in the ARF that is excited by the incident light.

3. Results and Discussion

The 1064-nm output pulse of the microlaser has a pulse duration of 2.0 ns (FWHM) with a measured $M^2 = 1.4$. The 1064-nm beam is focused to a $1/e^2$ waist diameter of 0.10 mm. As shown in Fig. 2(a), the measured frequency-doubling conversion efficiency is in close agreement with that predicted by SNLO. At the maximum 1064-nm input energy of 200 μJ, the measured conversion efficiency is 69%.

The seeding efficiency in the OPA is evaluated by two methods. First, we compare the unseeded 761-nm signal beam spectrum to that with the addition of the seed laser. Unseeded, the $LiTaO_3$ operates as an optical parametric generator, with the unseeded output signal beam spanning 0.5 nm (FWHM) as measured using the OSA. Upon seeding, the signal beam spectrally narrows to less than the 0.015-nm resolution limit of the OSA, and the out-of-band signal is suppressed by 23 dB. The ratio of the in-band to out-of-band pulse energy for the OPA is 28 dB. Second, the linewidth of the amplified 761-nm signal beam is measured using a 5-GHz free spectral range etalon to be 767 ± 23 MHz.

The amplified signal beam from the OPA is focused to a $1/e^2$ waist diameter of 0.19 mm within 1 mm of the input face of the first walkoff-compensating LBO crystal. The frequency-doubling conversion efficiency for this

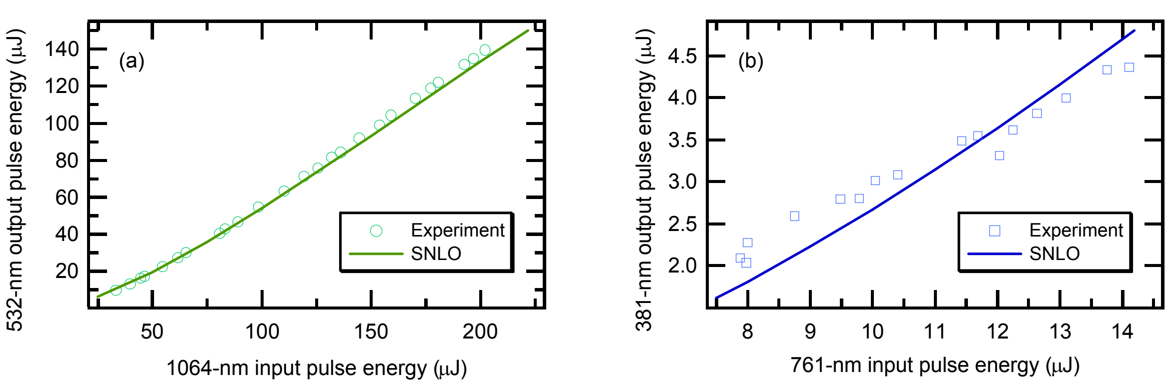

Fig. 2. Frequency doubling efficiency of (a) 1064 nm and (b) 761 nm.

stage is presented in Fig. 2(b) alongside that calculated with SNLO. A conversion efficiency of 31% is measured at the maximum 761-nm input energy of 14 μJ.

Lastly, the 761-nm and 381-nm beams are focused into the KDP crystal with a $1/e^2$ beam waist diameter of 0.13 mm. The frequency-conversion efficiency for this final stage is 63% below that predicted by SNLO. This disparity results from poor overlap between the 761-nm and 381-nm beams. However, for the maximum input energies measured, 13 μJ and 6.0 μJ of 761-nm and 381-nm light, respectively, the 254-nm output beam has a pulse energy of 1.8 μJ. Note that the required pulse energy is 0.1 μJ. Given that 254-nm pulse energies 18× that required are achieved and the need for laser compactness, we elected not to add additional optics to improve the spatial overlap of the 761-nm and 381-nm beams.

Using the observed characteristics of the Hg^0 laser system, we model the expected performance of the ARF. The transmission profile of the ARF is generated assuming a Voigt profile for the Hg-202 absorption lineshape, and the resulting transmission curves for several temperatures are shown in Fig. 3(a). In addition, the scattering from the laser and Hg^0 emission from the probe volume are presented in Fig. 3(a). The Hg^0 emission distribution is produced by convolving a Voigt profile for the Hg^0 absorption with the laser lineshape. A Gaussian spectral profile is assumed for the 254-nm output with a $\sqrt{3} \times 767$ MHz = 1.3-GHz linewidth (FWHM). As illustrated in Fig. 3(a), increasing the ARF temperature widens the absorption feature. Consequently, at higher temperatures the ARF will provide improved discrimination between the Hg^0 emission and narrower laser scatter. The improvement to the signal-to-background ratio (SBR) by using the ARF, defined as the ratio of the percent of transmitted Hg^0 emission to percent of transmitted laser scatter, is illustrated in Fig. 3(b).

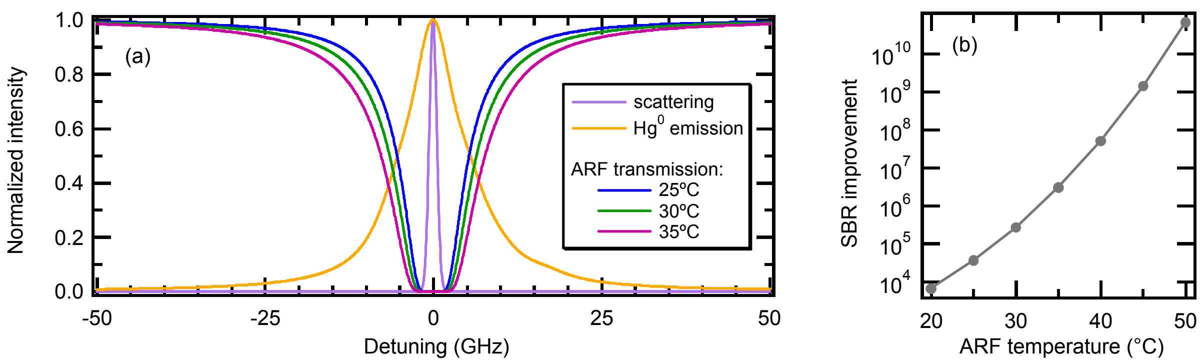

Fig. 3. (a) Spectral modeling of the laser scatter, Hg^0 emission from the probe volume, and ARF transmission for several temperatures. The probe volume collisional and Doppler widths are assumed to be 7.5 and 1.0 GHz, respectively. (b) Signal-to-background ratio improvement for increasing ARF temperatures.

4. Conclusions

We have designed a 254-nm laser source suitable for detecting Hg^0 emissions from coal-fired power plants with high sensitivity and specificity. The measured output pulse energy of the laser is 1.8 μJ, well exceeding the required level of 0.1 μJ. The laser's narrow linewidth enables the use of an atomic resonance filter to provide effective differentiation between the laser scatter and Hg^0 emission.

Continuing efforts in our laboratory are directed towards characterizing the linewidth of the 381-nm and 254-nm beams. In addition, the transmission of the ARF will be measured and compared to our simulations. Finally, the Nd:YAG microlaser will be replaced with a Sandia-built fiber amplifier operating at 13 kHz with a pulse energy of 110 μJ and a pulse duration of 840 ps. The higher repetition rate of this pump source is compatible with photon-counting detection and will yield significantly higher average power for the UV laser, thereby enabling sub-ppb detection limits for Hg^0.

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5. References

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