



# Laser Applications to Chemical, Security, and Environmental Analysis: introduction to the feature issue

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The eighteenth topical meeting on Laser Applications to Chemical, Security, and Environmental Analysis (LACSEA) was held in Vancouver, Canada from 11–15 July 2022, as part of the Optica Optical Sensors and Sensing Congress in a hybrid format allowing on-site and online attendance. The meeting featured a broad range of distinguished papers focusing on recent advances in laser and optical spectroscopy. A total of 52 contributed and invited papers were presented during the meeting, including topics such as photo-acoustic spectroscopy, imaging, non-linear technologies, frequency combs, remote sensing, environmental monitoring, aerosols, combustion diagnostics, hypersonic flow diagnostics, nuclear diagnostics, fs/ps applications, and machine learning and computational sensing. © 2023 Optica Publishing Group

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This feature issue of *Applied Optics* consists of 13 papers by experts in subject matters representing a wide range of interesting and novel aspects of the expanding research field, specifically, in the category of spectroscopy, materials and instrumentations.

Spectroscopic measurements were employed for characterization of rock samples and for the study of the weatherization of lithium hydride (LiH). Song *et al.* [1] used laser-induced breakdown spectroscopy (LIBS) in combination with a 1D convolutional neural network for automated rock classification. An improved Bayesian optimization algorithm was proposed to improve the efficiency of rock structure analysis. In comparison to other algorithms, the modelling time could be reduced by about 65%. Pinson *et al.* [2] studied environmental conditioning of lithium compounds using dual Raman LIBS and machine learning algorithms to analyze the spectra. It was also shown that machine learning can be used to model environmental conditions such as humidity and temperature derived from known LiH reactions.

Wang *et al.* [3] fabricated a high-performance Si/Ga<sub>2</sub>O<sub>3</sub>/CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> photodetectors using Ga<sub>2</sub>O<sub>3</sub> as electron-transport-layers. The Ga<sub>2</sub>O<sub>3</sub> based perovskite photodetectors achieved high performance including R value up to 7.2 mA W<sup>-1</sup> and D\* as high as 7.448 × 10<sup>10</sup> Jones, respectively and showed performance reproducibility and stability under

780 nm light illumination, indicating a good potential for future applications in photodetectors.

Hou *et al.* [4] constructed a CD/Cu-MOFs ratiometric fluorescence sensor based on the Förster resonance energy transfer (FRET) principle, which is highly sensitive for the detection of Hg<sup>2+</sup>. A green fluorescent coumarin derivative was obtained by a one-step condensation reaction with an excitation wavelength of 440 nm and an emission wavelength of 505 nm. A combination with a Cu-MOFs allowed the construction of a new type of CD/Cu-MOFs ratio fluorescence sensor for a highly selective detection of Hg<sup>2+</sup>. The fluorescence emission intensity of the system showed a good logarithmic relationship with the lowest detection limit reaching 3.76 × 10<sup>-9</sup> nM.

Nakagawa *et al.* [5] demonstrated a newly designed hollow optical fiber coupler for a mid-infrared laser heterodyne spectrometer. A high transmission efficiency of ~87%/m was achieved for a coherent laser source and ~89%/m for an incoherent blackbody source. The asymmetric branching ratio of the hollow fiber coupler was newly achieved, enabling efficient collection of signals from both the local oscillator and an observed target on one side of the output port of the coupler. A sensitivity performance suggested that the transmission of the coherent local oscillator laser through a hollow optical fiber has an almost equal sensitivity to that without a fiber.

Gilvey *et al.* [6] used quantum-cascade laser-based absorption-spectroscopy to measure temperature, pressure, and NO at 500 kHz in shock-heated air at elevated pressure. They demonstrated that the diagnostic is capable for measurements at high temperatures ( $\sim 5500$  K) and pressures (12 atm). The measurements were in excellent agreement with CEA predictions.

Sahoo *et al.* [7] demonstrated a novel approach for making direct quenching measurements at atmospheric conditions using nanosecond lasers. The quenching measurements were demonstrated in a krypton-perturber system and the  $5P[\frac{3}{2}]_2 \leftarrow 4p^{61}S_0$  two-photon electronic transition was accessed. The relative quenching rates for different perturber species (Air, CH<sub>4</sub>, C<sub>2</sub>H<sub>4</sub>, and CO<sub>2</sub>) are reported at 1 atm and 300 K. However, the present technique is limited to measurements of the relative quenching rate, and not able to measure absolute quenching rates, which have to be calculated from the relative quenching rates.

Elkhazraji *et al.* [8] used a widely tunable light source emitting in the long-wavelength mid-infrared region applied to chemical kinetics and environmental monitoring applications. The laser source was based on a difference-frequency-generation process in a nonlinear orientation-patterned GaAs crystal, resulting in tunable laser light between 11.58  $\mu\text{m}$  and 15.00  $\mu\text{m}$  in a quasi-cw manner. They developed detection schemes for benzene, as well as selective diagnostics tailored for HCN and toluene sensing in a shock-tube.

Gu *et al.* [9] studied the AP-HTPB propellant combustion under strain conditions with Laser absorption spectroscopy. Here, they measured the effects of static and dynamic strains on the burning rate, temperature, CO, and CO<sub>2</sub> formation of aluminized ammonium perchlorate (AP)-hydroxyl terminated poly-butadiene (HTPB) propellant combustion at pressures of 0.1 MPa, 0.2 MPa and 0.5 MPa. Strain was applied onto the solid propellant by exerting static and cyclic loadings. The propellant burning rate was acquired by a 4 kHz high-speed photography system, and the combustion temperature, CO, and CO<sub>2</sub> column densities were measured at 10 kHz through laser absorption spectroscopy. The propellant burning rate and combustion temperature are found to increase with higher relative strain and the generation of CO and CO<sub>2</sub> during AP-HTPB propellant combustion can be related to the high and low temperature chemical kinetics associated with AP-HTPB combustion.

Different novel approaches for velocimetry are also presented in the special issue, including that for hypersonic flows. Li *et al.* [10] proposed an adaptive Gaussian weighted integral-based water flow velocity measurement method to solve the processing problem of weak non-stationary signals in long-distance laser Doppler water flow velocimetry. The Kaiser self-multiplication window is designed to suppress spectral leakage for the asynchronously sampled data. Jiang *et al.* [11] demonstrated krypton tagging velocimetry (KTV) and picosecond laser electronic excitation tagging (PLEET) at a 100-kHz-rate in Mach 18 flow conditions employing a burst-mode laser system and a custom-made optical parametric oscillator. The measured freestream flow velocities from both KTV and PLEET agreed well with

theoretical calculations and the increase in repetition rate provided a better capability to perform time-resolved velocimetry measurements in hypersonic flow environments.

This issue includes two invited topical reviews that cover the recent progress in high-speed laser diagnostics for hypersonic flows and analytical spectroscopy for nuclear diagnostics. Rao *et al.* [12] summarized recent advances in machine learning in analytical spectroscopy for nuclear diagnostics. For diagnostic analysis of nuclear materials, LIBS, Raman spectroscopy and x-ray fluorescence spectroscopy have achieved considerable improvements over the past years. Especially when rapid analytical measurements of nuclear materials with no sample preparation are needed, spectroscopic methods can be beneficial in comparison to mass spectroscopic analysis. As in many other applications, a combination with machine learning implementations is beneficial. These techniques have been applied to nuclear forensics, nuclear fuel manufacturing and quality control, and general diagnostic analysis of nuclear materials. Handheld LIBS devices for example can be used for rapid chemical analysis of plutonium metal and can quickly quantify impurity elements such as silicon. Tabletop LIBS setups with high resolution spectrographs can instantly provide measurements of the gallium content with detection limits, as low as 60 ppm. Here, the implementation of ensembles of trees for regression analysis can yield high fidelity prediction models from complex LIBS spectra. LIBS and Raman can also be used in tandem for environmental aging studies of lithium compounds. specific environmental exposure conditions could be determined by relating them to changes in atomic and molecular emissions of various lithium compounds. Jiang *et al.* [13] reviewed the recent progress in high speed laser diagnostics for hypersonic flows. Although high-speed laser diagnostics has achieved significant progress in the last 15 years, the diagnostic techniques for hypersonic flows are still limited to a few techniques due to the numerous challenges associated with hypersonic flows, facilities, and available equipment. The review covers the progress of two main laser diagnostics techniques at repetition rates of 100 kHz and above: focused laser differential interferometry (FLDI) and pulse-burst laser-based diagnostic (PBLD). FLDI diagnostics require a CW laser, photodetector and a few optics and thus are less expensive. Although they are limited by single- or multi-point measurements, the FLDI technique has been widely applied to hypersonic flows. PBLD is a powerful technique capable of molecular tagging velocimetry, planar laser-induced fluorescence, and PLEET with detailed multiple flow parameter measurements in 1D-4D. However, its application is limited to a few groups. Here, the currently available high-speed cameras are a limiting factor for high-repetition rate 2D measurements, due to the limited pixel or limited frame numbers at 100s of kHz or MHz rates, which are essential for high spatial resolution and longtime recording, as well as due to their high cost.

Over the years the LACSEA meeting has become one of the premier must-attend meetings in the field of laser-based spectroscopic analytics, especially for applications associated with combustion diagnostics, chemical analysis, environmental monitoring, and trace species detection related to security and explosives surveillance. Consequently, the next LACSEA meeting is already being planned and we invite you to participate in

this stimulating experience and to contribute with your recent results.

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