REMOTE IMAGING LASER-INDUCED BREAKDOWN SPECTROSCOPY AND REMOTE ABLATIVE CLEANING

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ABSTRACT

Remote laser-induced breakdown spectroscopy (LIBS) measurements have been performed using the mobile lidar system of Lund Institute of Technology. Radiation from a frequency-tripled Nd:YAG laser was focused on the remote target using a specially designed beam expander, placed co-axially with the receiving telescope. The beam path of both the transmitting and receiving telescope are folded using a computer-controlled mirror, enabling scanning of the laser on the target and thus performing imaging.

1. INTRODUCTION

Laser-induced breakdown spectroscopy (LIBS) is a method for elemental analysis [1]. A laser beam is focused on the target to create a small plasma, in which free, excited, atoms and ions are created. As the plasma cools, the atoms and ions relax, emitting species-specific radiation. When this spectrum is detected, a characteristic signal is achieved that can serve as a fingerprint of the target material.

LIBS is widely used for elemental analysis in laboratory environments. It is known as a quasi-non-destructive analysis method; the surface of the target is naturally damaged, but the amount of ablated material can be very small and still make measurements possible without causing visible damage to the target. Information on the constituent elements in the sample is obtained, but there is no information on the composition. With a database of known samples, the relative amounts of the constituents can be found, but this information cannot be deduced immediately from one spectrum.

Remote LIBS measurements have been demonstrated in early Russian work [2] and recently both with nano-second pulses [3] and by femto-second pulses forming filaments [4]. We have performed what we believe to be the first remote imaging LIBS and the results of these measurements have recently been published [5,6].

2. MEASUREMENT SET-UP

The Lund lidar system [7] is a fully mobile optical parametric oscillator-based differential absorption lidar system, but it is also well suited for measurements on solid targets, e.g. laser-induced fluorescence or remote LIBS. An overview of the system is shown in Fig. 1.



Fig. 1. The mobile lidar system used in the measurements.

For remote LIBS, the laser radiation has to be focused to create a spark at the remote target. To create a focus at a large distance, the beam has to be expanded at the source. It is also important to create an intense plasma to be able to detect the signal remotely.

Laser radiation from a frequency-tripled Nd:YAG laser is guided through a beam expander where the distances between the negative and the two positive lenses can be adjusted to place the focus at the target. The expanded laser beam is sent through a dome at the roof of the lidar truck, where a computer-controllable folding mirror is used to direct the radiation to the desired spot.

Clearly, if the target is farther away, it will be more difficult to focus the laser. This can to some extent be compensated by increasing the pulse energy, but in our case, this is limited by the damage tolerance of the transmitting optics.

When a spark is created at the target, the excited atoms and ions re-emit light as they relax. The created light is collected using a 40-cm-diameter Newtonian telescope and focused onto the tip of an optical fiber connected to a spectrometer. The transmitting and receiving optical axes are the same, which makes imaging easy as the detected light comes from the direction in which the excitation light was transmitted. The spectrum in the wavelength range 280-810 nm can be detected. The detector is gated to focus on the interesting signal. The gate can be adjusted, both its start and duration, which makes it possible to detect only the interesting signal and to study the temporal development of the plasma radiation.

The signal is averaged over a number of shots, to reduce the noise in the measurement. When the signal has been detected, the beam direction is changed and the signal from the next point is detected, and so on. In this way, a scan over an area is achieved, i.e. by whiskbroom imaging.

3. MEASUREMENTS

The measurements were performed in Lund with the system docked to the Physics Department building (see Fig. 2) but could as well have been performed anywhere, as the system is fully mobile, housed in a Volvo F610 truck. Nano-second laser pulses with 20 Hz frequency at 355 nm from a tripled Nd:YAG laser with an energy of about 150 mJ was used. The laser pulse was expanded and the beam expander was adjusted to create a focus on the target at 60 m distance.



Fig. 2. The measurement area. The lidar instrumentation is housed in the truck seen on the left and the target area is on the roof of a house 60 m away. The lower left corner shows a close-up of the target area.

As target, metal plates of different materials mounted on a screen were used. The materials used were copper, stainless steel, brass, aluminum and iron. A spark was created and the signal was detected. Imaging could be performed by scanning the laser beam over the area.

Measurements were performed changing delay and gate of the detector, but on the same target point, to check the temporal development of the plasma radiation. Measurements were also performed on minerals and stones.

Also, tests on remote ablative cleaning were performed. An area on an Italian garden statue that had been exposed to weather and wind was successfully cleaned by scanning the laser beam over the area. The process was self-terminating, so only the contamination was ablated and the underlying substance was unaffected.

4. **RESULTS**

An example of a LIBS spectrum from a brass plate is seen in Fig. 3. The spectrum is taken with 60 m distance from the source and detector to the target. The laser pulse energy was 170 mJ and the spectrum was averaged over 500 laser shots. In the spectrum, lines from copper and zinc can be clearly seen. Also some sodium contamination is noted.



Fig. 3. A LIBS spectrum from brass, taken from 60 m distance

When performing a scan, each point gives a spectrum, specific to the target material. By analyzing the shape of the spectra, classification can be accomplished.

A vertical scan over the metal plates was performed. The data can be analyzed in different ways. A simple way would be to monitor the intensity at a dominant spectral line, but this is sensitive to overlapping features and can thus give erroneous results. A better way would be to make a linear correlation with a known spectrum and thus take the shape of the entire spectrum into consideration.

Also, the data can be analyzed by principal component analysis. The data are transformed onto a new set of base directions, principal components (PCs), which are set as an orthogonal system such that as much of the variation as possible is described by the first PC, as much as possible of the remaining variance in the second one, and so on. The different materials could easily be separated, as seen in Fig. 4, where the score along PC1 has been plotted against PC3. It is also noted that copper and brass are close to each other and that iron and stainless steel are close to each other. This is expected, they have similar spectra since they partly contain the same substances.

The measurements to study the temporal development of the spectrum showed, as expected, that at early times the white-light continuum dominates and later the spectral lines becomes more dominating.



Fig. 4. Principal component analysis of the spectra in the vertical scan, PC1 against PC3.

Fig. 5 shows a calcite stone, which contains some quartz and some clay minerals. A spark could be created on the stone and a part of the detected LIBS spectrum can be seen in Fig. 6. Calcium lines are clearly seen as well as silicon lines from the quartz and aluminum from the clay minerals.



Fig. 5. A calcite stone.



Fig. 6. A part of the spectrum from the calcite stone.

Fig. 7 shows a part of an Italian garden statue after cleaning of an area $(6\times4 \text{ cm}^2)$ on the statue. The cleaning has been performed at 60 m range by scanning the laser beam over the area. The arrow indicates the cleaned area. As the laser beam is steered by using stepper motors, only certain points on the target can be reached. To be able to clean the entire area, the laser beam is rapidly moved over the area several times, thus cleaning all parts of the area.



Fig. 7. A cleaned area on an Italian garden statue.

For remote laser cleaning, the goal is to remove all the contamination and keep all the underlying material. This can be automated in two main ways. First, there is the scheme we have performed, to keep the laser fluence optimal – such that the contamination is removed, but not the material. This is then a self-terminating process, it does not matter how long we fire the laser on the already clean material, it will not be affected. However, this method may not be very efficient, since very high laser fluences cannot be used, and hence the time for cleaning the object will be long. On the other hand, the threshold fluence is also wavelength dependent, so a clever choice of wavelength might make it possible to use high fluences to efficiently remove the contamination while yet not destroying the material.

The other alternative is spectroscopic control. The LIBS spectrum is studied while the treatment is performed. When the spectrum changes to the appearance of the clean substance, the treatment is stopped. Then all the contamination is ablated, but as soon as the last contamination disappears, the measurement is halted. However, for our situation, the signal strength was not enough to detect the spectrum while cleaning.

5. CONCLUSIONS

Our results show that remote LIBS can be used for assessment of materials, even in imaging mode, where thematic maps can be produced. The same system can also be used for remote laser cleaning.

6. OUTLOOK

We believe that there is great potential in these types of measurements, with application towards the cultural heritage sector. Previous studies from our group have shown that fluorescence lidar is an efficient method for the assessment of e.g. building façades [8-10]. These measurements could be complemented by LIBS measurements to gain even more information. The two techniques could be integrated in the same system, as both the fluorescence and the LIBS measurements that have been performed have been done with the same system, even though the system is initially intended for DIAL. Clearly, this means that the measurements can be improved.

Also, the same system could be used for laser cleaning of the building façades. Laser cleaning performed today is usually expensive (due to required scaffolding), causes dangerous working environment (due to ablated particles and gases being inhaled) and can cause problems due to uncomfortable working positions. Using remote laser cleaning, these problems are eliminated. However, only the parts which can be reached by a laser beam can be cleaned, but then again, these are the same parts that can be seen by a spectator on the ground. Spectroscopic control of the ablation can also be integrated.

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