Detection and Mapping of Latent Fingerprints by Laser-Induced Breakdown Spectroscopy

M. T. TASCHUK,* Y. Y. TSUI, and R. FEDOSEJEVS
University of Alberta, Electrical and Computer Engineering, Edmonton, AB T6G 2V4, Canada

Detection of latent fingerprints on a Si wafer by laser-induced breakdown spectroscopy (LIBS) is demonstrated using approximately 120 fs pulses at 400 nm with energies of 84 ± 7 μJ. The presence of a fingerprint ridge is found by observing the Na emission lines from the transferred skin oil. The presence of the thin layer of transferred oil was also found to be sufficient to suppress the LIBS signal from the Si substrate, giving an alternative method of mapping the latent fingerprint using the Si emission. A two-dimensional image of a latent fingerprint can be successfully collected using these techniques.

Index Headings: Laser-induced breakdown spectroscopy; LIBS; Latent fingerprint detection; Two-dimensional surface mapping.

INTRODUCTION

Laser-induced breakdown spectroscopy (LIBS) is a rapid and flexible material characterization technique. The LIBS technique uses a tightly focused laser beam to break down the surface of a sample, creating a plasma formed of the elemental constituents of the target material. As this plasma cools it emits electromagnetic line radiation at wavelengths characteristic of the individual atomic constituents of the original target. Recent studies have extended the range of LIBS to much lower pulse energies and smaller focal spots than those traditionally used.1–9 This new regime, referred to as micro-LIBS or μLIBS, utilizes pulse energies below 100 μJ and allows the probing of smaller sample spots, which maximizes the spatial resolution for two-dimensional (2D) mapping of surfaces.1–6,9

Fingerprints are a highly reliable method of identifying humans, as they have been found to be unique to each individual.10 Objects that have been touched often have transferred fingerprints that can be used in criminal investigations or security applications. Fingerprints found at crime scenes are classified into three types: visible prints such as those made when an ink-coated finger touches a surface and leaves a print, plastic prints, which are a mechanical impression in soft materials such as wax, and latent prints, which result from a transfer of oils from the finger to a surface.10 The traditional method for detecting and collecting latent fingerprints is to lightly dust a suspect surface with fingerprint powder, which will tend to stick to the latent fingerprint. The dusted print can then be lifted from the surface with an adhesive plastic sheet for later evaluation.10

Detection of fingerprints using lasers was first demonstrated by Dalrymple et al., who excited the fluorescence of fingerprint oils using an argon ion laser.11 The major challenges in fingerprint detection with laser techniques are low emission levels from the fingerprint oils and interference from the fluorescent signal from the background substrate. Strategies for improving the emission signal from fingerprint oils include selection of laser wavelength,12 as well as chemical treatments to tag the fingerprint for enhanced fluorescence response.13,14

Elimination of background interference has been the focus of much work in this area.15–18 Hooker et al. used a 0.3 m f/4 monochromator to spectrally resolve the fluorescence to optimize the choice of fluorescent dye agents and filters for photography of fingerprints.15 An alternative technique is to use gated acquisition of the fluorescent fingerprint image to avoid ambient light. Work by Wenchong et al. uses intensified charged-coupled device (ICCD) detection to eliminate contributions from highly illuminated substrates such as exterior walls during daylight.16 Recent work by Dinish et al.17,18 makes use of phase-resolved fluorescence from fingerprint samples. This technique modulates the illumination laser at several MHz to induce a phase shift between the substrate background and the latent fingerprint signals that can be used to detect the print.

The LIBS technique offers an alternative to the traditional optical methods used to date for fingerprint detection as it directly detects the elemental signature of latent fingerprints. In many cases, the substrate on which the latent fingerprint is found will have only trace amounts of the elements found in the fingerprint. Recent work by Lopez-Moreno et al. has demonstrated detection of human fingerprints at a distance of 30 m using the fundamental of a Nd:YAG with 350 mJ laser pulses.19 The fingerprints used as samples for their work were composed of the natural skin oils, various explosive residues, and other samples. Successful discrimination between fingerprints of different compositions was achieved.

In the current paper, a preliminary investigation of the capabilities of μLIBS not only for detection but also for imaging of latent fingerprints is reported. Lower pulse energies are required to minimize damage to the fingerprint during the mapping process. As shown in the present results, with low-energy femtosecond pulses the sensitivity depth is reduced to a depth comparable to the fingerprint film. This effect reduces the effect of the substrate in fingerprint detection and imaging. Characteristics of the elemental optical emission of latent fingerprint on a Si substrate have been measured and are presented below.

EXPERIMENTAL SETUP

The experimental setup used for LIBS fingerprint detection and mapping is given in Fig. 1. A Ti:Sapphire (Spectra-Physics Hurricane) laser, which produces approximately 120 fs (full-width at half-maximum, FWHM) at 800 nm pulses, is frequency doubled using a lithium triborate (LBO) crystal. The doubled beam is at 400 nm and gives a maximum pulse energy of approximately 90 μJ on target. The pulse width of the doubled beam is estimated at approximately 120 fs (FWHM) on target. A total of four dielectric 400 nm mirrors at 45 degrees are used to separate the fundamental and second
The spectral purity of the excitation beam was measured before the focal lens with a calibrated photodiode and an OG570 glass filter (Schott Glass) to block the 400 nm laser light. The residual 800 nm laser light energy was approximately 40 nJ/pulse when the 400 nm light was approximately 90 nJ/pulse for a spectral contrast ratio of about 2200. This level of 800 nm leakage light is not expected to be a significant factor for the experiments presented here, especially considering that the 800 nm light will be out of focus for the conditions used here.

The pulse energy was controlled with a combination of a half-wave plate and Glan–Taylor prism in the fundamental beam before the doubling operation. A photodiode has been calibrated for pulse energy delivered to the target plane using the power meter. The precision of the calibration factor is 6% and the absolute error of the power meter is 5%. For the laser pulse energies reported in this paper, the absolute error is estimated at approximately 8%. The laser pulse has a Gaussian spatial profile, and was focused to a 9 ± 1 μm (FWHM) spot on the sample surface using a 10 cm focal length lens. For an 80 μJ pulse, the peak irradiance on the target is approximately 3 × 10^{13} W cm^{-2}.

The LIBS emission was measured using a 1/4 m, f/3.9 imaging spectrometer (Oriel MS260i) combined with an ICCD. A grating with 600 lines mm^{-1} and a blaze angle of 400 nm was used, giving a reciprocal linear dispersion of 0.168 nm channel^{-1} or 6.5 pm μm^{-1}. The entrance slit of the spectrometer was 100 μm for all experiments. A spectral calibration of the spectrometer was carried out using a Hg lamp (Oriel 6035) as a wavelength standard. The Hg lamp calibration was combined with the wavelength calibration provided by the manufacturer for the work presented here.

The spectra were recorded using an ICCD (Andor iStar DH720–25 mm). Due to the very low pulse energies used in μLIBS, very short delay times are necessary to achieve an optimum signal-to-noise ratio (SNR). The delay time was
calibrated using unfocused laser light scattered from a barium sulfate diffuser plate placed at the target site. A single 84 μJ pulse at 400 nm with a gate delay of approximately 5 ns and a gate width of 1 μs.

Both spectra were generated with a single 84 μJ pulse at 400 nm with a gate delay of approximately 5 ns and a gate width of 1 μs. A gate delay of approximately 5 ns was chosen for the initial studies presented here. The gate width used was 1 μs, which is long enough to capture all available emission without accumulating significant detector noise for the gain conditions used here.

In the case of 2D mapping of surfaces, it is necessary to extract as much information as possible from each shot, as the surface properties change after the first laser pulse. In previous work, it has been shown that it is possible to estimate SNR from a single shot using neighboring regions of the spectrum that are spectrally quiet. For an initial analysis of the results presented here, a single-channel definition of SNR is used:

$$\text{SNR} = \frac{I_{\text{peak}}}{\sigma_{\text{background}}}$$

where $I_{\text{peak}}$ is the background-corrected strength of the line of interest in one channel and $\sigma_{\text{background}}$ is the standard deviation of a spectrally quiet region away from the peak, once background correction has been performed. A linear model was fit to and subtracted from these regions to account for the general trend of the background.

The fingerprints used as samples in this paper were taken from the right thumb of one of the authors. First, a Si substrate was rinsed with ethanol and wiped clean with lens paper prior to the deposition of the fingerprint. The fingerprints were prepared by brushing the thumb against the forehead and then lightly rolling the ball of the thumb against a cleaned Si substrate. The preparation of suitable fingerprints is a challenge: too much pressure smears the fingerprint, reducing definition; too little pressure yields an incomplete fingerprint. Only fingerprints that looked complete and were clearly visible in the reflected image of fluorescent room lights on a Si wafer were used for the experiments described below.

**EXPERIMENTAL RESULTS**

Initial trials for fingerprint detection by LIBS were undertaken using a fingerprint left on a Si wafer. It is expected that the oils that are transferred to surfaces in latent fingerprints contain Na, which is a good candidate for LIBS detection using the doublet line at 589.0 nm and 589.6 nm. Silicon provides a smooth surface for the latent fingerprint and has strong emission in the observed spectral region. For each shot, Na and Si SNR were calculated using Eq. 1. In the work presented here, two spectrally quiet regions were used for background corrections: the first from 570.4 nm to 573.7 nm, and the second from 592.2 nm to 595.5 nm. These background regions are used for both Na and Si signals. In the case of Na, the peak wavelength used was 589.2 nm, and for Si the peak wavelength used was 576.2 nm. The wavelengths quoted for the background and peak regions correspond to channel centers and thus do not necessarily agree with handbook values for observed lines.

The Si line observed here is identified as the 288.16 nm line in the second order. This was confirmed by placing a GG395 glass filter (Schott Glass) in front of the slit, which strongly filters wavelengths below 380 nm. This test removed the Si signal entirely, ruling out the possibility that the 575.4 nm, 576.3 nm, or 577.2 nm neutral Si lines contributed to the Si signal observed here. Figure 2 gives sample spectra from a fingerprint ridge (Fig. 2a), and from a region between fingerprint ridges (Fig. 2b). Both spectra are single-shot acquisitions taken with 84 μJ pulses at 400 nm. These spectra indicate that the presence of a fingerprint ridge can be sufficient to shield the substrate from the laser pulse, as no Si signal is observed in Fig. 2a. In this case, a fingerprint ridge must be thick enough that the part of the ablated layer that contributes to the LIBS signal does not penetrate into the Si substrate.

A one-dimensional (1D) LIBS scan of a latent fingerprint sample is given in Fig. 3. A step size of approximately 50 μm was used for a 100 shot series along the surface for a total mapped distance of 5 mm for all scans. For each location on the target, SNR was calculated for both Na and Si. Both signals are periodic in space with a period of approximately 750 μm, but out of phase. Si signals are lower in regions where a strong Na signal was obtained, suggesting that fingerprint ridges are sufficient to suppress the LIBS signal from the Si substrate.

As a further investigation of the suppression of the Si signal, a Si wafer with a latent fingerprint was rinsed with ethanol and wiped with lens paper: the ethanol was dripped onto the substrate and the lens paper was gently drawn over the substrate. This procedure was performed twice, and a 1D LIBS scan was made of the cleaned region. However, as can be seen in Fig. 4a, this procedure has left Na on the wafer at concentrations that were still detectable by LIBS. The washing procedure was repeated twice more, for a total of four wash-and-wipe cycles. Another 1D LIBS scan of the surface was
made, and the average Na SNR was below the 99% confidence detection limit (Fig. 4b). However, Na is still detected at a few locations in the 1D scan, indicating that fragments of the print have survived the cleaning process. In both the twice-cleaned sample and the quadruple-cleaned sample, Si was visible at all locations, and at an average SNR of approximately 30. These signal levels are somewhat consistent with those observed in the 1D fingerprint scan presented in Fig. 3b, indicating that the fingerprints prepared here do not cover the entire surface and that the laser pulse is interacting with a mostly bare Si substrate between fingerprint ridges.

A 2D scan of a 1 mm by 5 mm region of a latent fingerprint from the right thumb of one of the authors was performed using laser pulses with energies of 84 μJ. The spatial step size was approximately 50 μm in both dimensions. SNR for Na and Si as a function of position are presented in Fig. 5. The distribution of Na SNR values has a small number of outliers with SNR values above 60. In order to highlight the mapping capability of this technique, the 2D map of Na SNR presented in Fig. 5a is limited to a maximum SNR of 50. Ridges of the fingerprint are visible in the Na image (Fig. 5a), and gaps between ridges are visible in the Si image (Fig. 5b).

Using optical microscopy, re-deposited material is visible surrounding the mapped area. This effect has been the subject of some study in the LIBS literature, and in principle may affect the quality of any mapping process performed by LIBS. It is known that in some cases the shock wave launched by the LIBS ablation process can clean the re-deposited material such that a fresh surface is available for 2D mapping. In the current case, the clarity of the fingerprint maps presented in Fig. 5 suggest that re-deposition did not significantly affect the results presented here. However, further investigation of the degree to which re-deposited material can affect the mapping of fingerprints for both the current sample type and other substrate materials will be required to quantify the effect.

FIG. 3. 1D scan of a latent fingerprint on a Si wafer using single-shot 84 μJ pulses at 400 nm and approximately 50 μm spacing between shots. (a) Na signals and (b) Si signals are periodic in space with a period of approximately 750 μm. Si signal levels are lower where a strong Na signal is observed, suggesting that the presence of a fingerprint is sufficient to suppress the LIBS signal for the Si substrate.

FIG. 4. 1D LIBS scans of control samples. Samples were imprinted with a fingerprint as described in the text. Samples were then rinsed with ethanol and wiped with lens paper. (a) After repeating the cleaning procedure twice, Na was still detected at reduced levels, though fingerprint structure had been destroyed. (b) After a total of four cycles of the cleaning procedure, Na is no longer reliably detectable on average. On both the twice-washed and quadruple-washed sample (c) Si is visible at all spatial locations at SNR levels similar to those observed in the gaps in the fingerprint observed in Fig. 3b.

DISCUSSION

While detection and mapping of a latent fingerprint using 400 nm 84 μJ, 120 fs laser pulses has been demonstrated, many issues remain. These include the time required to acquire a fingerprint image, the level of damage to the latent fingerprint, difficulty in preparing consistent fingerprint samples, the effect of re-deposition of ablated material during a mapping process, and the detection of reliable fingerprint signal on different surface materials.

The time taken to acquire the image presented in Fig. 5 is approximately 20 minutes of laser firing at 2 Hz. This image represents approximately 1% of a total thumb print area of approximately 6 cm. The acquisition time could be improved significantly by moving to the multi-kHz repetition rates possible with fiber or microchip lasers. Significant changes to the experimental setup for controlling direction of the laser beam and acquisition of the signals would be required. It is expected that with acousto-optic or galvano-metric deflectors...
and a photo-multiplier tube, data acquisition would be possible at kHz repetition rates, which would allow the acquisition of a full print in a reasonable time. In addition, the step size used here for preliminary testing may be unnecessarily small for fingerprint mapping.

Another difficulty is the possibility of unacceptable damage to the fingerprint. LIBS is generally considered a nondestructive technique, but in the case of small structures that are similar in scale size to the probe spot this is no longer the case. In the current scans, there is significant damage to the print, though the overall structure remains visible. At pulse energies of approximately 80 lJ crater diameters are approximately 50 µm. If step size were increased to 100 µm, only approximately 25% of the print would be damaged by the LIBS mapping process. Further, with even lower energies, it may be possible to further reduce the damage to the print to the point that a substantial fraction of the fingerprint survives the LIBS measurement process. Further work will be required to ascertain the compatibility of the LIBS fingerprint scanning process with the requirements of law enforcement and security applications.

Additional complications arise from the preparation of the latent fingerprint itself, which is a difficult standardization challenge. As a result, it is not certain that the conditions used here are optimal for the overall distribution of latent fingerprints that might be observed using this technique. Additional optimization opportunities exist in the choice of wavelength.\textsuperscript{12} Use of the third harmonic of the Ti:Sapphire laser at 266 nm may reduce the energies necessary to produce good SNR due to increased absorption in the fingerprint oils.

Re-deposition of ablated material was observed for the current 2D mapping process. Based on the quality of the fingerprint image acquired in the present work, it is not believed that the re-deposited material is a limiting factor in the acquisition of fingerprint images. However, further work will be required to quantify this effect. In addition, to test the utility of the technique, fingerprint samples on a variety of different substrates will need to be investigated.

Despite the difficulties listed above, this technique has potential to provide an alternative to the current fluorescent fingerprint imaging techniques. While the Si substrates used here do not contribute any Na emission to the signal, other substrate materials may contribute emission in the Na band. However, in those cases where Na is found in the substrate, it may be possible to make use of the substrate signal suppression effect observed here to distinguish between Na emission from a latent fingerprint and contributions from the substrate. By observing other elemental lines from the substrate, the substrate signal suppression effect may help identify regions covered by fingerprint ridges. However, further work will be required to ascertain the feasibility of this approach.

Gornushkin et al. and Freedman et al. have demonstrated that microchip lasers can be used to obtain useful LIBS data.\textsuperscript{7,8} With further optimization of the fingerprint detection operating parameters and the expected increases in the output of microchip lasers, it may be possible to design a portable device capable of detection and mapping of fingerprints. By increasing the shot spacing to 200 µm and the repetition rate to approximately 5 kHz, which would be possible with a microchip laser, a 6 cm\textsuperscript{2} fingerprint could be imaged in
approximately 3 seconds. Such a device may reduce the time required to process a crime scene by providing near real time feedback to forensic investigators.

CONCLUSION

A preliminary investigation of the capabilities of femtosecond µLIBS for detection and mapping of latent fingerprints has been conducted. Detection and mapping of latent fingerprint structure has been demonstrated using 400 nm, 84 µJ pulses. The presence of a latent fingerprint has been shown to suppress the line emission from the Si substrate in regions covered with a fingerprint ridge, indicating that the sensitivity depth for femtosecond pulses is similar to the thickness of the fingerprint. The substrate signal suppression effect may make it possible to detect the fingerprint pattern by monitoring the suppression of other substrate emission lines rather than the Na emission. A 2D image of a 1 mm by 5 mm region of a fingerprint has been successfully collected. Further study is expected to reduce the energy requirements for fingerprint detection. A multi-kHz laser, high-speed scanners, and improved data acquisition should reduce the current time required to acquire a fingerprint image significantly.

ACKNOWLEDGMENTS

The authors gratefully acknowledge financial support for this research from both MPB Technologies Inc. and the Natural Sciences and Engineering Research Council of Canada.