MICRO-RAMAN MAPPING of DISSIMILAR INKS on PAPER

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Impact

This presentation demonstrates the use of micro Raman mapping as a visual technique well suited for courtroom presentations.

Abstract

Attendees will be presented with an example of identifying the order of ink deposition where lines cross by micro-Raman mapping. Two types of maps will be presented. Surface maps that indicate the locations of individual inks on the paper surface and depth maps that indicate ink layer structure orthogonal to the paper surface. Maps appear as photographic images with ink locations presented in false colors. This approach produces a four step visual interpretation of the analytical data that is easy for juries to understand. The steps are: 1. Identify the Inks in the Document. 2. Date Acquisition from Questioned Area. 3 Constructing a Map of Ink Locations. 4. Top Ink Determination at the Point of Line Crossing by Surface and Depth Mapping.

Introduction

When documents are altered the order of ink layer deposition where lines cross is an important determination confronting questioned document examiners. Knowledge of the top layer suggests the intent of the alternation while the lower layer provides information about the document's original content. Together, this information helps establish motive, and may suggest a suspect or timeline of the crime.

Materials and Methods

Instrument Settings
In general a trial-and-error approach is required to determine the optimal compromise between spatial resolution and signal strength. Raman excitation was 785nm, 30 second exposure time, 10% laser power and two accumulations. Spectral response was measured from 3200cm^{-1} to 850cm^{-1}. Different magnification and confocality settings were needed for each step.

Step 1. Reference spectra were determined at low confocality and 20x magnification providing a full field of view in high image and high signal strength.

Step 2 and **Step 4.** 50x magnification was needed to produce 20 micrometer x 20 micrometer pixel dimensions for mapping. Data was acquired at low confocality to maximize signal strength and overcome minor out-of-focus conditions caused by paper surface undulations.

Step 5, depth mapping was performed using 50x at high confocality. Additional acquisitions were required to compensate for low signal strength.

Spectra Matching
Identifying different inks and mapping their locations relied on matching the spectra of unknown inks in the questioned area to known reference spectra of the inks in the body of the document. Direct Classical Least Squares (DCLS) fitting of the unknown data to a linear combination of the known ink reference spectra was conducted using the "Component Analysis" feature in WiRE 3.4 software (1).

Results

Step 1. Identify Inks in the Document Ink Reference Spectra and False Color Assignment

Conventional ballpoint inks were tested by making a figure of a cross with single lines of different ink. Figure 1 presents an image of a test sample showing reference spectra for Ink A and Ink B that were assigned false colors red and green respectively.
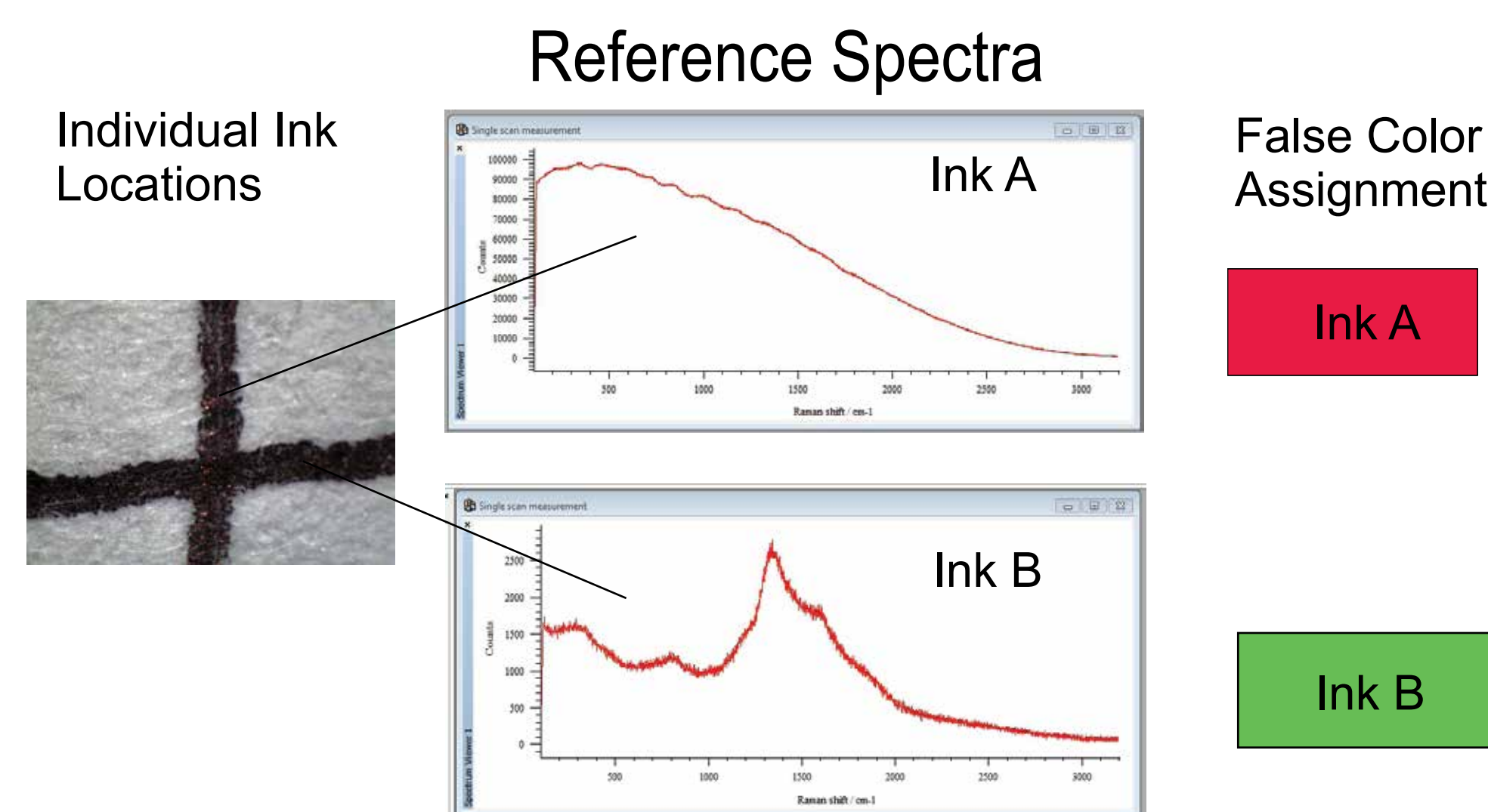
Reference Spectra

Individual Ink Locations

Ink A

Ink B

False Color Assignment

Ink A

Ink B

Figure 1. Reference Spectra For Ink A (red) and Ink B (green).

Step 2. Data Acquisition from the Questioned Area, False Color Shows Locations of Individual Inks

Digital photographs are a collection of pixels arranged in a grid pattern in which each pixel contains a dot of photographic data. Like a photographic image, each pixel in a spectral map contains photographic data plus the infrared spectrum at that location. To determine which ink was present, the spectrum at each pixel location was compared to the ink reference spectra using DCLS component comparison in Wire 3.4 software (1).
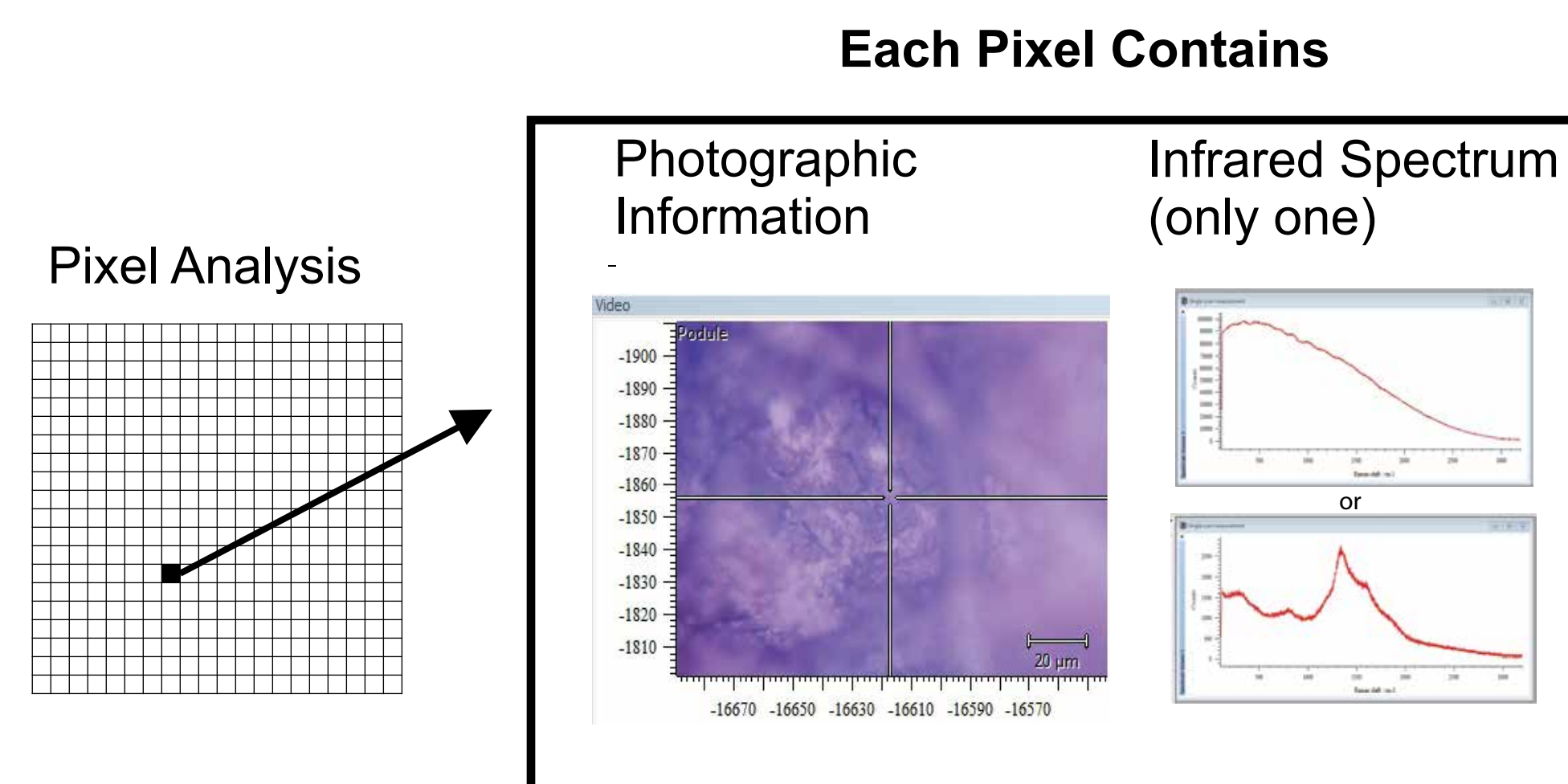

Each Pixel Contains

Pixel Analysis

Photographic Information

Infrared Spectrum (only one)

or

Figure 2. Analyzing Each Pixel Location to Determine Photographic and Infrared Spectral Information.

Step 3. Constructing a Map of Ink Locations

When creating the spectral map of the questioned area the pixel was drawn in the color assigned to the ink spectrum associated with that location. In Figure 3 false colors indicate positions of the inks.
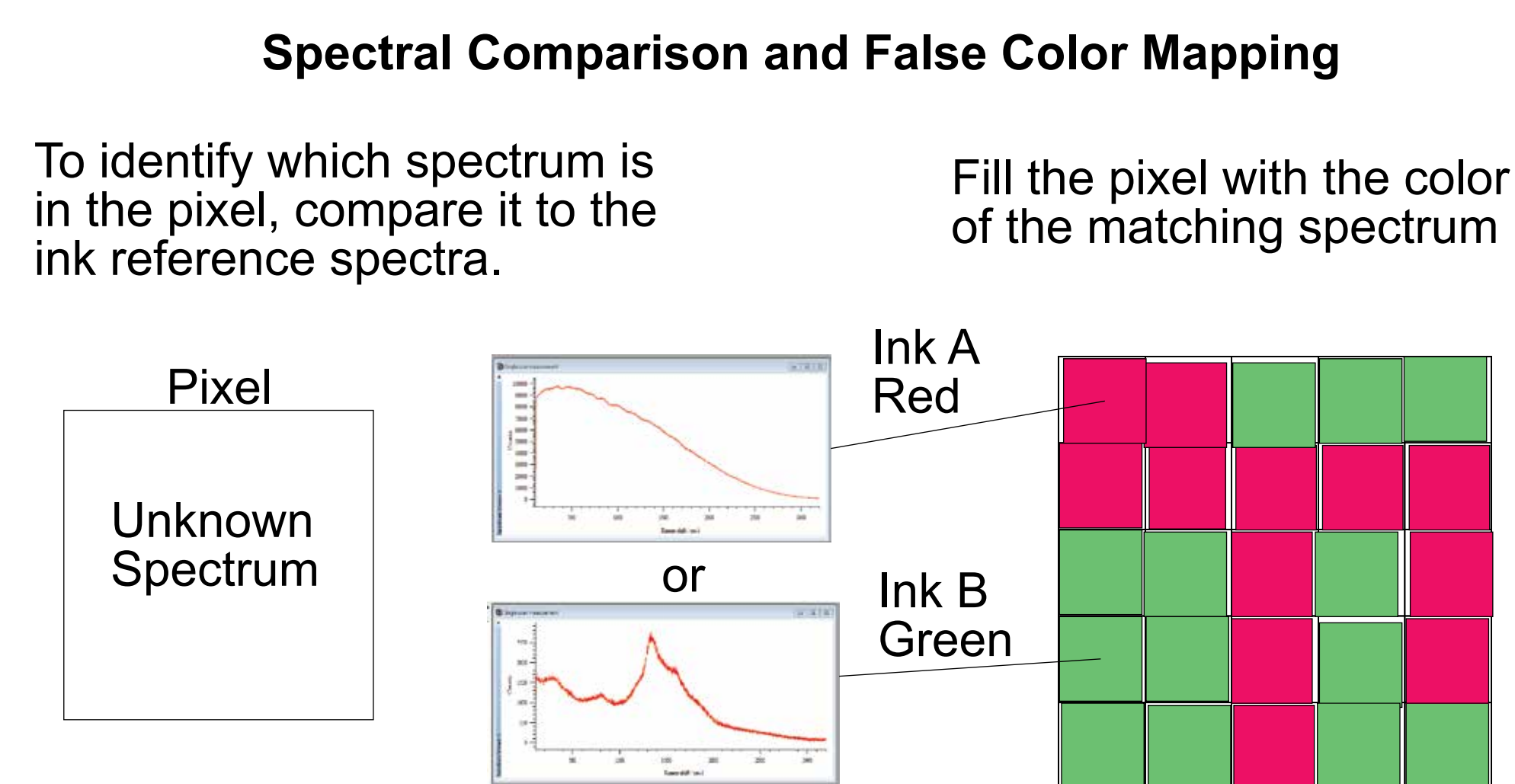
Spectral Comparison and False Color Mapping

To identify which spectrum is in the pixel, compare it to the ink reference spectra.

Fill the pixel with the color of the matching spectrum

Pixel

Unknown Spectrum

or

Ink A Red

Ink B Green

Figure 3. Compiling the Infrared Spectral Information in Each Pixel to Create a False Color Map Showing the Location of Inks A and B

Figure 4 presents an example of spectral mapping applied to document alternation. The date 2001 was written in ink B then changed to 2004 with ink A. Because ink B was assigned the false color of green each pixel is rendered in green when the spectral map is drawn. Likewise, the false color red indicates alternations made with ink A. In this example, the sequence in which the original writing and subsequent alternation was evident from the context of the writing and determining the top ink layer at the point of crossing was unnecessary.
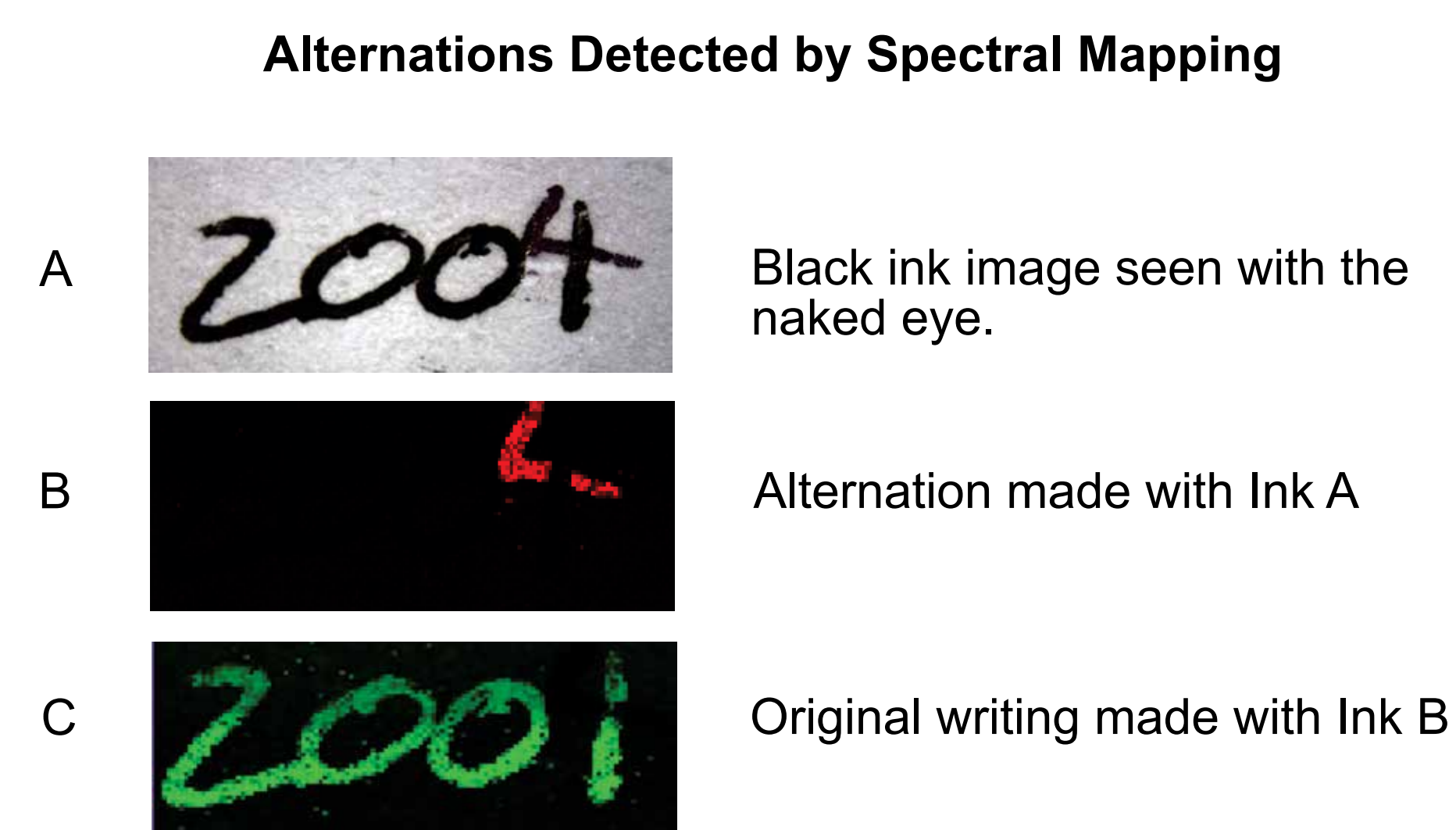
Alternations Detected by Spectral Mapping

A — Black ink image seen with the naked eye.

B — Alternation made with Ink A

C — Original writing made with Ink B

Figure 4. An Example of False Color Mapping in Document Alternation

Step 4. Top Ink Determination at the Point of Line Crossing

In circumstances where the sequence of deposition is not evident from the context of the document, additional analysis is necessary to determine the sequence of application. Both surface and depth mapping techniques provide this information in different ways.

Surface Mapping Technique
In figure 5, the grid in the light colored box defines the mapped area; cells in the grid are pixels. Pixel area was set by the motorized stage "Step" setting that adjusts the distance traveled between data acquisition points. Greater detail can be achieved with smaller steps and higher magnification at the expense of reduced signal strength. Trial-and-error was needed to determine the optimal balance of these settings.
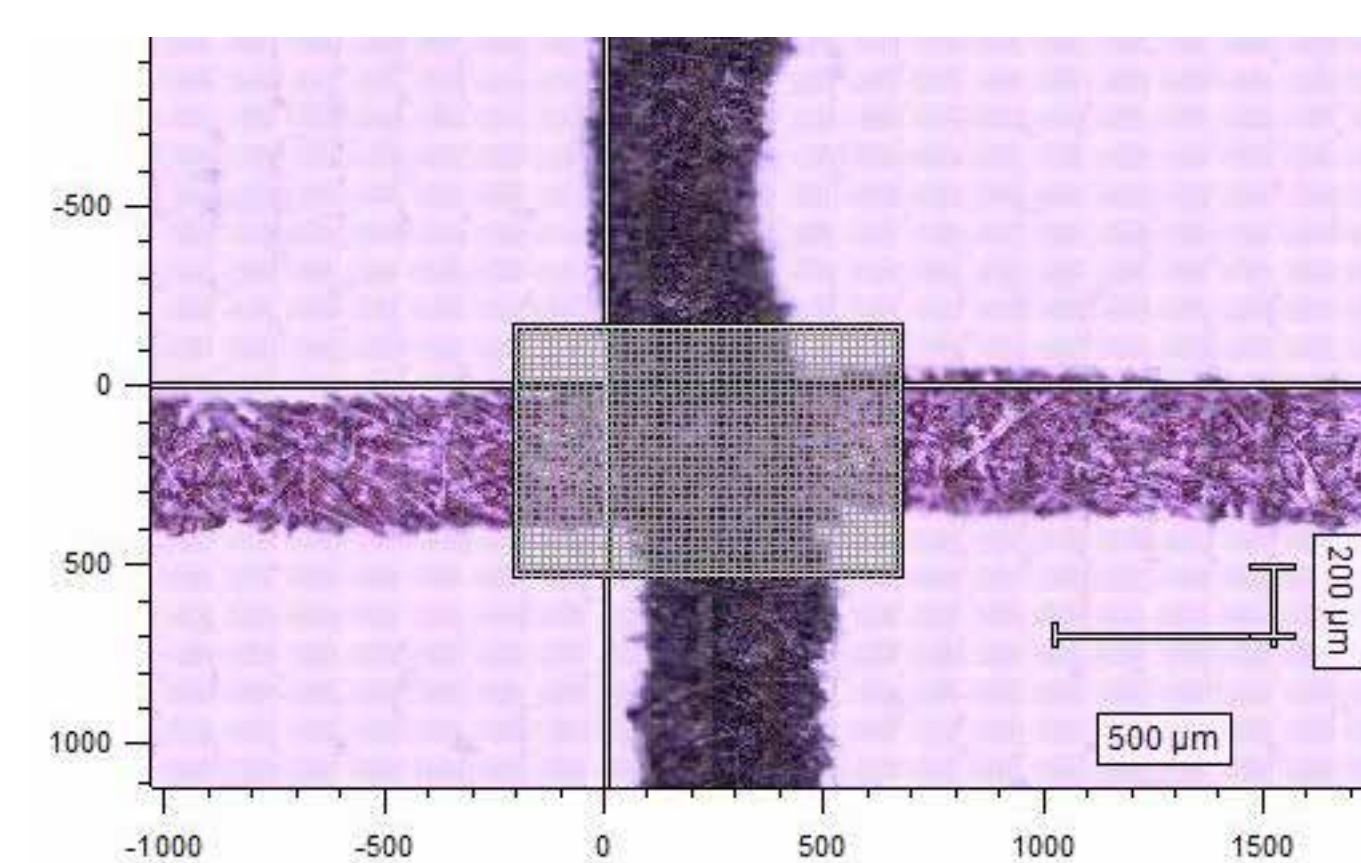
Figure 5. Mapping at the Point of Line Crossing
The light colored grid marks the mapped area while cells within the grid are individual pixels.

At the point of line crossing in Figure 6, ink locations were visualized in false color. When a single ink dominated, it was interpreted as the top layer and understood to be the adulteration. In the top panel the predominately red colored map indicates that Ink A was on top; supported by red continuing uninterrupted outside of the crossing area. Although Ink B was present outside of the crossing area its intensity inside the crossing area was greatly diminished supporting the conclusion that Ink B was the underlying layer.
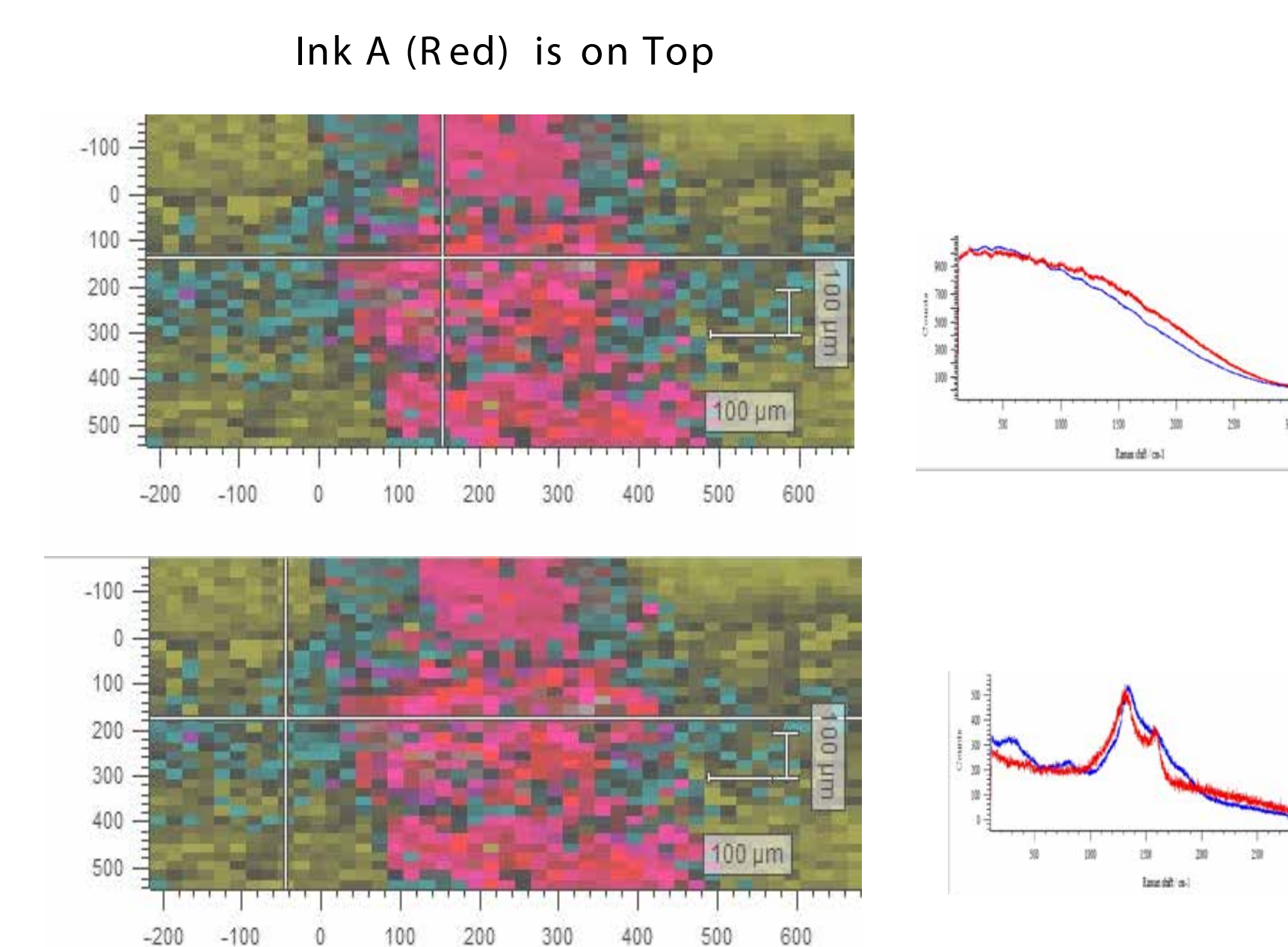
Ink A (Red) is on Top

Figure 6. Example of Surfacing where Lines Cross
Ink identity was determined by the spectra (right) taken from the location marked by the cross hairs in the photographs on the left. Based on its dominence at the point of crossing and continuing uninterrupted past the crossing boundaries, Ink A was determined to be the top layer.

Depth Mapping Technique
Depth maps depend on the ability of a confocal microscope to confine its focus on thin layers that are not affected by adjacent layers. Additionally, by focusing above the specimen and repeatedly lowering the microscopic objective the "in focus" layer visualizes a vertical stack of thin layers. Infrared energy has the ability to penetrate past the surface of the specimen into the underlying material. Using this approach micrometer thick layers were analyzed starting above the ink layers and progressing into the paper. When approached from above the first ink layer is the alteration and the underlying layer is the original writing. In Figure 7, the paper plane is located at Z= O micrometers ---negative values were below the paper surface while positive values were above. The vertical axis is signal intensity where large values indicate the presence of ink. Inks were identified by their spectra and indicated by their false color as before. In Figure 7A the uppermost layer, identified as ink A, was detected at 40 micrometer while the signal for ink B was at its lowest intensity. At the paper surface in Figure 7B, the signal intensity for ink B was near its highest level while the signal for Ink A had fallen to near its minimum. The interpretation is ink A was the uppermost layer representing the alternation while Ink B was the underlying layer representing the original writing. Figure 5D shows this layering in false color.
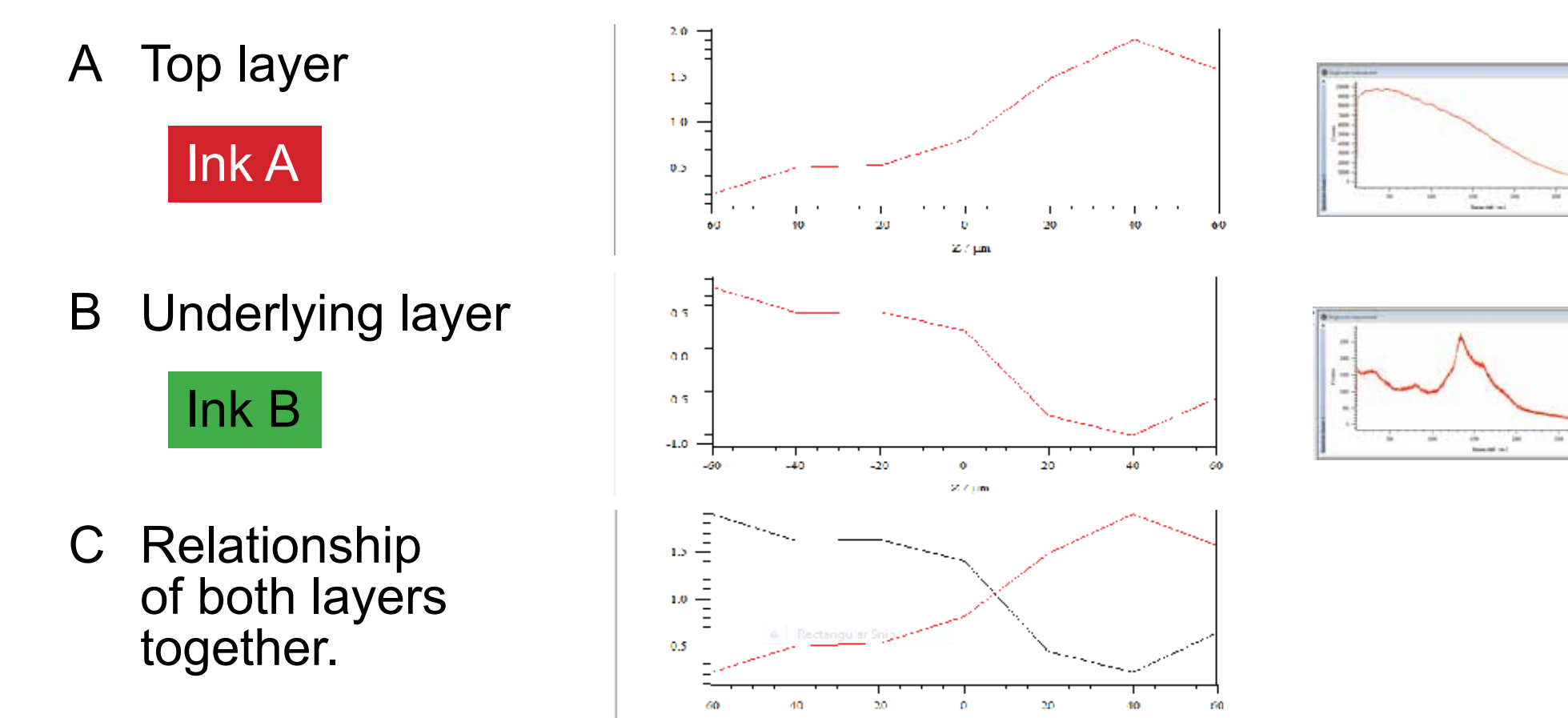
Depth Map Showing Ink Layers

A — Top layer — Ink A

B — Underlying layer — Ink B

C — Relationship of both layers together.

Figure 6. Depth Map Indicating the Sequence of Line Deposition
In panel A above the intensity of both ink spectra are plotted against distance above the paper surface followed by the individual plots for the top and underlying layers in panels B and C respectively. The inks were identified by their spectra using the "Component DCLS" function to identify the best fit of the pixel spectrum to the ink reference specta.
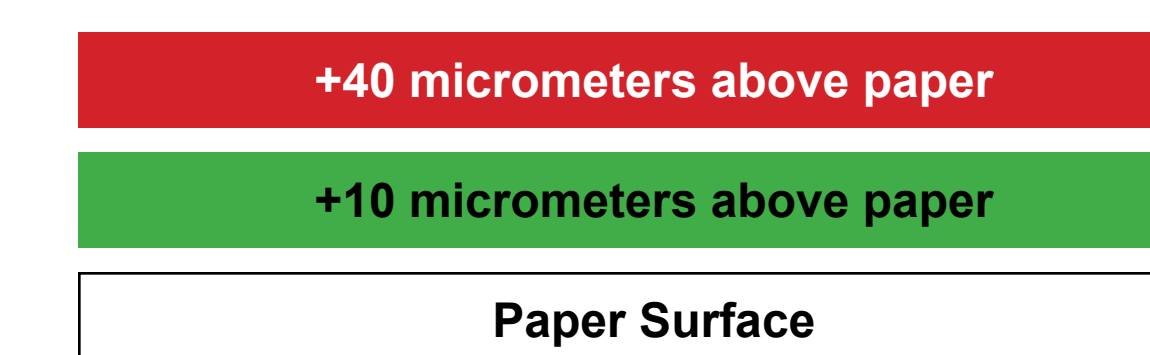
+40 micrometers above paper

+10 micrometers above paper

Paper Surface

Figure 7. Summary of Depth Mapping Data

Discussion

Although technical details are critical to the examiner, they often overwhelm or confuse the jury. This work illustrates a visual presentation based on instrumental analysis that requires little technical knowledge to be understood by a lay audience.

References

WiRE 3.4 spectrometer software is a produce of Renishaw plc, Spectroscopy Products Division, Old Town, Wotton-under-Edge, Gloucestershire, GL12 7DW, United Kingdom

Acknowledgements