Characterization of European Lacquers by Terahertz (THz) Reflectometric Imaging

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***Abstract*—In this study a European lacquerware replica has been investigated by terahertz (THz) reflectometric imaging. The inspected lacquerware is a wooden panel covered by multiple complex layers of lacquers and plaster. Utilizing pulsed Terahertz Time-Domain Imaging (THz-TDI) in reflection mode, we observe non-invasively buried layers of the lacquerware replica, including the internal structure of the wooden panel itself. We find that non-invasive terahertz reflectometric imaging analysis of lacquerware is can provide conservators with important information about the condition of the compositional layers, potentially aiding in the development of appropriate conservation treatments. With the same technique we have performed a surface material mapping. The material distribution has been enhanced through reflected THz composite RGB false color rendering, where RGB mapping allows distinction between different materials and textures on the surface of the lacquerware. The contrast between different textures is enabled by wavelength-dependent scattering from the surface, as well as differences in the composition of the surface layer.**

Keywords—terahertz; laquerware; cultural heritage; non-invasive; false-color; tomography

I. INTRODUCTION

A. The Significance of Terahertz Time-Domain Imaging Applied to Lacquerwares Investigation

Lacquerwares, ornamental objects decorated by multiple, composite layers of lacquers, are one of the most significant expressions of Eastern Asian art. Deeply appreciated by Western countries, they had a large influence on the world's arts and crafts. Their production dates back to 5000 BCE in Asia and, starting from the sixteenth, they have been imported, collected and, finally, imitated in Europe. Techniques of production and materials used vary a lot depending on lacquered objects historical and geographical context. Being composite objects, the preservation of lacquerwares is particularly complex [1].

Scientific analysis of lacquers could provide conservators with crucial information about composition and condition of the constituent layers, aiding in the development of appropriate conservation treatments. The knowledge of the inner structure, stratigraphy and condition of the subsurface layers of these multi-layer objects is, in this sense, highly relevant in conservation when stability problems such as delamination or internal cracking are considered. The standard methods for visualize the inner structure of artifacts are infrared reflectography and X-ray radiography, techniques that are usually complemented with microscopic analysis of cross-sectioned samples. These methods have limitations, since the mentioned imaging techniques provide only flat, two-dimensional (2D) images of three-dimensional (3D) structured systems, while cross sections require invasive sampling that provides only local information, with the risk of not being representative of the object as a whole.

Terahertz time-domain imaging (THz-TDI) in reflection geometry, being capable of highlight interfaces between layers in a stratigraphic buildup, could be a complementary technique for obtaining structural information about lacquered objects. Unlike X-ray radiography and infrared reflectography THz-TDI provides not only 2-D images but also subsurface 3-D images and unlike cross-sectioned samples it provides stratigraphic images (b-scans) contactless and non-invasively. Furthermore, like the other standard imaging techniques it could be used to identify promising sample sites that are not otherwise detectable with additional advantages: terahertz radiation is non-ionizing due to the low photon energy (1 THz ~ 4.1 meV) and has very low power levels (approximately 1 microwatt), so that the inner structure may be visualized without adverse effects on the artifact [2, 3].

B. Terahertz Time-Domain Imaging in Reflection Geometry: Fundamentals

Terahertz time-domain imaging (or terahertz pulsed imaging) is a noninvasive, coherent imaging technique that can acquire inherently 3-D data. Terahertz radiation (1 THz = 10^{12} Hz) lies between the millimeter and infrared regions of the electromagnetic spectrum. It is typically defined as the frequency range of 0.1-10 THz (0.3-3 mm in wavelength), but THz-TDI systems typically have a usable frequency range of approximately 0.1-3 THz. In relation with imaging, the main attractive property of terahertz radiation is its ability to penetrate optically opaque and non-conducting materials.

For imaging applications, the source of contrast is the optical density of materials and in reflection geometry the back-reflected THz pulse is analyzed for reflections originating from the various interfaces present between the various sample layers. Specifically, terahertz refractive index

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mismatches result in reflections from buried layers and terahertz images are formed by the detection of absorption and refractive index changes, which in general occurs when there is a structural or material change in the sample under investigation. THz-TD systems emit and detect a very short electromagnetic pulse (of the order of less than a picosecond duration), with frequency content in the terahertz frequency range. The recorded quantity is the electrical field strength of the electromagnetic pulse as a function of time. In the majority of systems in use, the terahertz beam is raster scanned across an object over two spatial dimensions *x-y* and a time-domain pulse is recorded at each spatial coordinate in order to form the image.

The temporal spacing between the reflections is proportional to the optical thickness of the layers. In this sense, the method works in a manner similar to that of acoustic echo (ultrasound) equipment or radar depth measurements, which means that the time scale provides information about the penetrated depth of the signal that is reflected, enabling 3-D time-of-flight (ToF) imaging. The sign and amplitude of the reflected pulses provide information about the THz refractive index contrast between the different layers. The conversion of the pulses from the time to frequency domain by means of Fourier transformation could allow substance identification through chemical mapping, given that many solids exhibit characteristic spectral features in the 10 GHz - 4 THz frequency range. Both phase and amplitude information of the reflected signal can be obtained, enabling the investigation of both morphological and chemical changes. The structural and chemical information are convolved in a single reflected pulse and can only be separated if some properties of the sample are known *a priori*. Furthermore, in reflection geometry, THz-TDI measurements are strongly influenced by the scattering of the THz-waves caused by irregularity or grain-like structure of surfaces [4, 5]. Recently, an image processing technique for the visualization of buried layers through uneven surfaces has been reported and the method is in progress [6].

Terahertz images can be plotted utilizing different parameters. In time domain images can be reconstructed from measured data using the already mentioned pulse delay with regard to a reference pulse (ToF). Plan-type images (c-scans) can be displayed using the temporal amplitude (E(t)) of a pulse (either the value of the electrical field in the maximum, in the minimum, or the difference of maximum and minimum), the integration of a pulse or calculated power over a specific time interval (time window) and the centroid- or weighted peak-time. The frequency domain can also be exploited, using the spectral amplitude (E(ω)) at a specific frequency or frequency range, but also the amplitude or power integrated over a specific frequency range [7].

The lateral resolution (or spatial resolution) of Terahertz time-domain imaging system is less than 1000 micrometer (µm). It is a function of wavelength, and the beam waist diameter of the focused beam is limited by diffraction effects to a diameter approximately that of the wavelength (Abbes criteria). The reduction of the focal diameter is limited by the wavelength of THz radiation utilized. The ability to resolve closely spaced reflections (bandwidth-limited axial resolution or depth resolution) is determined by the temporal duration of the THz pulses. In terms of the spectral bandwidth of the terahertz pulse, the depth resolution is given by half of the coherence length of the radiation according to $L_C = c/\Delta\omega$, where $\Delta\omega$ is the spectral bandwidth and c is the speed of light in the intervening medium. Even if the resolution depth is limited by the spectral bandwidth of the terahertz radiation, different techniques have been recently used to improve the depth resolution [8, 9].

II. MATERIALS AND METHODS

A. *Analytical Instrumentation and Methodologies*

Terahertz time domain spectroscopic imaging (THz-TDSI) has been performed with a commercial (Picometrix T-Ray 4000) device, consisting of a femtosecond fiber laser coupled with 5-m long umbilical cords to a photoconductive terahertz-transceiver head mounted on an XY-scanning stage. The commercial system utilizes a pump-probe-system for generating and detecting short pulses of electromagnetic radiation at terahertz frequencies and utilizes a Labview-based operating system. A schematic representation of the device at normal incidence reflection geometry is shown in Fig. 1.

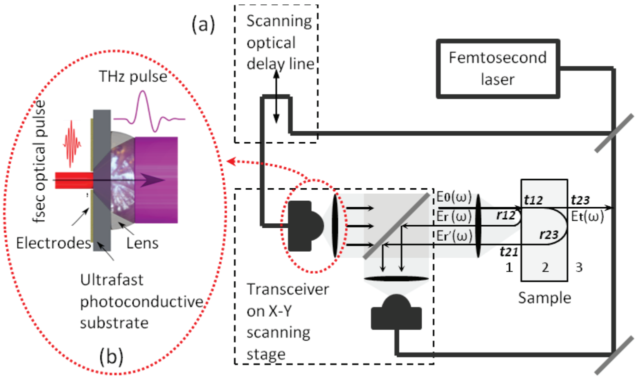

Fig. 1. (a) Schematic representation of the THz-TDI device used at normal-incidence reflection geometry. $E_0(\omega)$, incident radiation; $E_r(\omega)$ radiation reflected at the first interface; $E_r'(\omega)$, radiation reflected at the second interface; t_{12}, t_{23}, r_{12}, and r_{23}, complex Fresnel field transmission (t_{nn}) or reflection (r_{nn}) coefficients for the different media 1 (air), 2 (homogeneous sample), 3 (air). (b) Photoconductive switch for generation of terahertz pulses. The ultrafast laser pulse is impinged onto a biased semiconductor surface. The metallic electrodes supply the bias field to the photoconductive gap between the electrodes. The resulting current transients generate the THz pulses and THz radiation is collected into a collimated beam by a substrate lens attached to the structure

The emitted radiation is focused onto the sample by a polyethylene lens. The beam travels through air and is then detected and processed for frequency content after being reflected from the sample. The incident and the reflected beam are separated through a beam splitter. The frequency range covered is approximately 0.05 – 2 THz (wavelengths 6 – 0.15 mm, wavenumbers 1.7 – 66 cm^{-1}). The signal-to-noise-ratio (SNR) is about 80 dB at 0.5 THz. The lateral resolution (or spatial resolution) of the system is lower than 1000 micrometer (µm), while the bandwidth-limited axial resolution (or depth resolution), which is related to the dielectric properties of the investigated materials, is in the order of 20 micrometer (µm) [10]. The laquerware replica has been scanned in 0.5 mm steps, using a 320 ps measurement window, a time increment of 0.078 ps (4096 data points in each time trace) and a reflection configuration at normal incidence.

B. The Lacquerware Replica

The investigated European lacquerware (Fig. 2(a)) belongs to The Royal Danish Academy of Fine Arts (School of Architecture, Design and Conservation - Copenhagen, Denmark). The replica is a decorative work made of a wood panel covered with several layers of plaster and lacquers to depict floral and zoomorphic motifs. The wooden panel (layer I) is covered by a thin layer of lacquer (layer II) and a second layer of plaster (layer III) on which it has been applied a thick glossy black lacquer (layer IV). Engravings have been made on this surface and then marked out with Dragon's Blood (layer V) to trace the drawing of the figurative bas-reliefs realized with plaster (layer VI) inlaid with mother of pearl and beads. A lacquer layer coats the reliefs (VII), to hold silver-like glitters, which are then sealed under a layer of yellow colored lacquer (VIII), an imitation of gold.

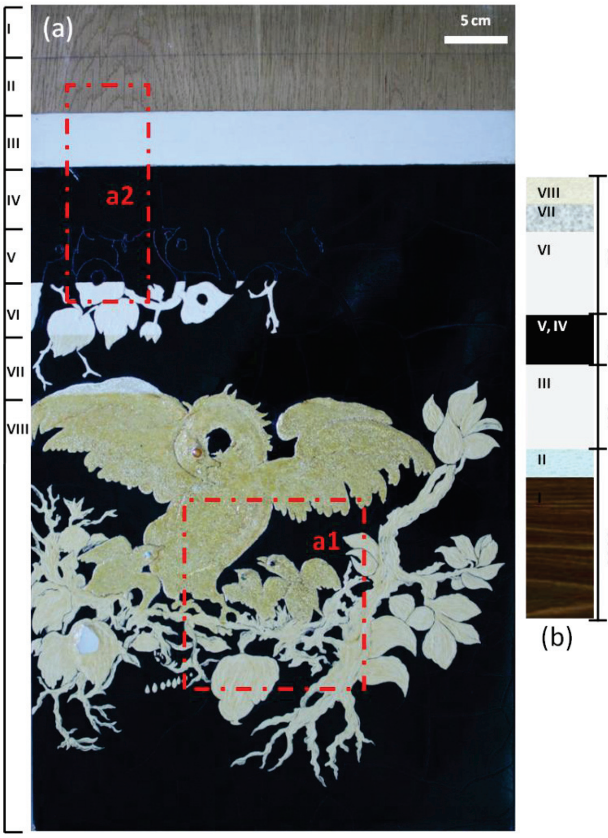

Fig. 2. (a) Visible-light photograph of the European lacquerware replica; the red lines outline the scanned areas (a1, a2). (b) Schematic stratigraphy of the lacquerware, with the indications of the average thicknesses measured.

III. RESULTS AND DISCUSSION

A. Plan-type Terahertz Reflection Images: C-scans

The value of the electric field measured for each spatial coordinate (x,y) of the scanned areas has been used for the bi-dimensional visualization of the lacquerware. A Matlab routine was coded to generate both frequency and time domain parametric terahertz images, shown in Fig. 3 for the scanned area a1 of Fig. 2(a). Using the parameter maximum of temporal amplitude (Fig. 3(a)), the information displayed arise predominantly from the first interface (air/ surface), so that the grain of the wood behind the surface are not as visible as using the parameter spectral amplitude integrated over the whole frequency range (Fig. 3(b)).

Contrast and detail enhancement has been obtained through histogram modifications (Fig. 3(d) and Fig. 3(e)), realized applying well developed image processing techniques such as logarithmic transformation, contrast-limited adaptive histogram equalization or gamma correction [11].

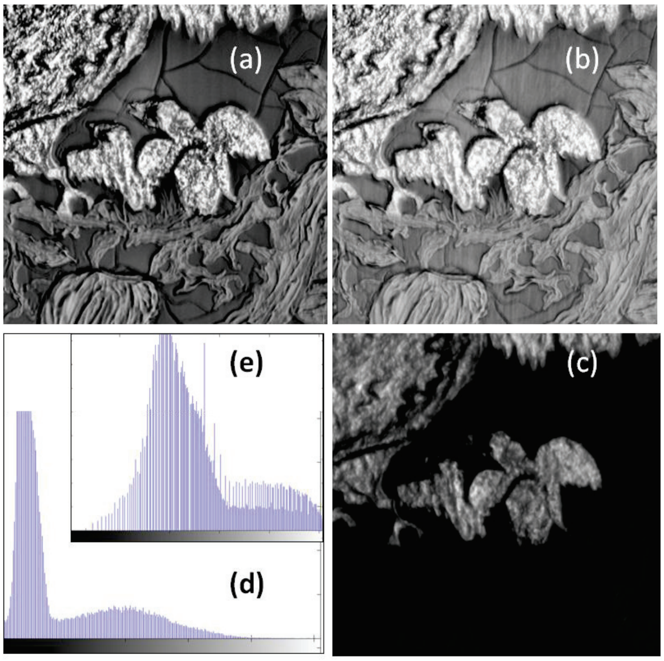

Fig. 3. (a)Terahertz time domain parametric image (maximum of temporal amplitude) after logarithmic transformation, contrast-limited adaptive histogram equalization and gamma correction. (b) Terahertz frequency domain image (integral of the spectral amplitude over the whole frequency range) after logarithmic transformation, contrast-limited adaptive histogram equalization and gamma correction. The location of the scanned area is indicated in Fig. 2(a). (c) Terahertz time domain parametric image (maximum of temporal amplitude) before logarithmic transformation, contrast-limited adaptive histogram equalization and gamma correction. (d) Histogram of Fig. 3(c). (e) Histogram of Fig. 3(b).

The terahertz reflection images in Fig. 3 are displayed by assigning a single intensity value for each pixel, resulting in different shades of gray from black to white proportional to the value of the parameter used (grayscale or intensity images). Another displaying method used in terahertz imaging application is pseudocoloring (some time referred as false-color), consisting in assigning arbitrary colors to the gray levels of an intensity image through a pseudocolor scale (or color map), where high pixel values are assigned one color (for example, red), and low pixel values are assigned another color (for example, blue), with other colors assigned to intermediate values (pseudocolor mapping) [12].

All the terahertz reflection images show a good terahertz optical contrast among the materials (black lacquer of the

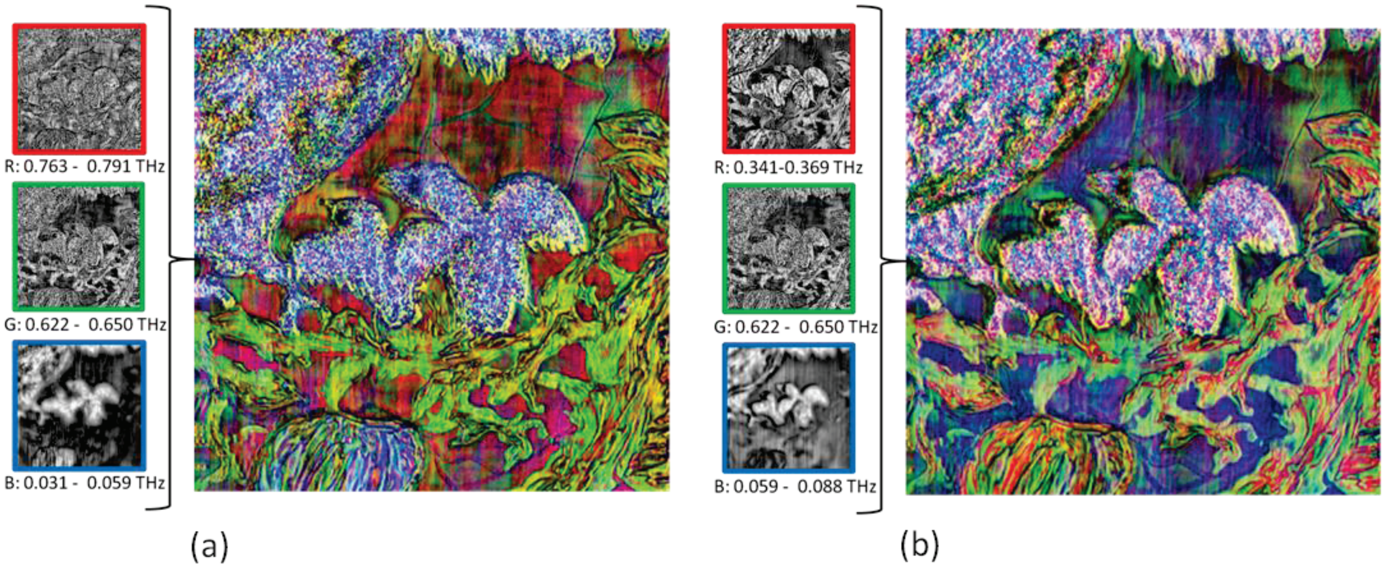

Fig. 4. (a) Terahertz reflection composite image obtained inserting the spectral amplitude integrated over the 0,0763 – 0,791 THz range image in the red channel, the spectral amplitude integrated over the 0,622 – 0,650 THz range image in the green channel and the spectral amplitude integrated over the 0,0031 – 0,059 THz range image in the blue channel. (b) Terahertz reflection composite image obtained inserting the spectral amplitude integrated over the 0,341 -0,369 THz range image in the red channel, the spectral amplitude integrated over the 0,622 – 0,650 THz range image in the green channel and the spectral amplitude integrated over the 0,059-0,088 THz range image in the blue channel.

background, plaster of the floral reliefs, plaster covered by lacquer and glitters of the animal reliefs). To improve this contrast, we have utilized composite RGB false color rendering to create reflected THz composite images. To the best of our knowledge this is the first application of this color rendering technique to terahertz reflection images. The 0.031 - 0.819 THz spectral window (9.67 - 0.819 mm in wavelength), characterized for having an acceptable signal to noise ratio, has been divided into twenty-five different frequency intervals (bands) and the integral of the spectral amplitude over each frequency interval has been calculated and used to form the images. All the possible permutations of sets of three images have been arranged into the RGB channels, obtaining the composite images. Two of them are shown in Fig. 4. Putting the three different bands together in one color composite gives a better visual impression of terahertz reflectivity contrast than displaying one band at a time or a unique band (whole frequency window). RGB false color rendering allows distinction between different materials and textures on the surface of the lacquerware. The contrast between different textures is enabled by wavelength-dependent scattering from the surface, as well as differences in the composition of the surface layer. An improvement is particularly evident for the animal reliefs, where reading of the dotted-like texture given by the metallic glitter inclusions in the lacquer is much clearer, thanks to the different RGB color of the metal grains with respect to the adjacent areas.

B. Plan-type Terahertz Reflection Images: B-scans

B-scans (non-invasive cross section images) have been realized by displaying the time-of-flight (travel time) of the electric field along the vertical axis and the (x,y) linear position of the transceiver along the horizontal axis.

Fig. 5(a) shows a b-scan image obtained from the scan-line dashed in red on the terahertz time domain parametric image of Fig. 5(b), showing the area a1 of Fig. 1(a). Six main interfaces have been found from the b-scan: I1 (air/wood interface), I2 (air/layer III interface), I3 (air/layer IV interface), I4 (air/layer VI interface), I5 (layer IV/layer III interface), I6 (layer III/wood interface). Additional minor interfaces are the ones formed because of inhomogeneities inside the wood and plaster layers. Notice that the wood grain is clearly visible in the lower part of the b-scan image.

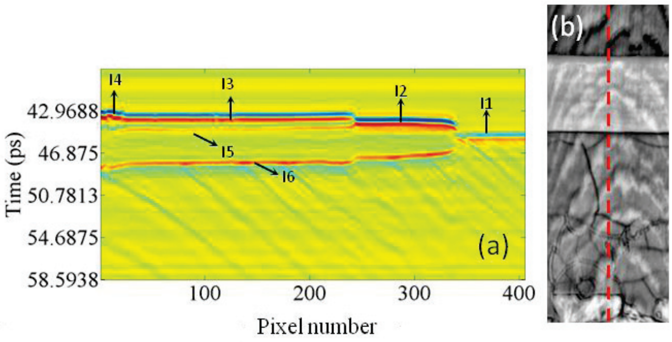

Fig. 5. (a) b-scan image obtained from the scan-line dashed in red on the terahertz time domain parametric image of (b). The six main interfaces found are I1 (air/wood interface), I2 (air/layer III interface), I3 (air/layer IV interface), I4 (air/layer VI interface), I5 (layer IV/layer III interface), I6 (layer III/wood interface). (b) Terahertz reflection image of the power of the electric field integrated over the whole time window; the dashed red line represent the scanline used to plot the b-scan of Fig. 5(a).

C. Time-of-Flight Plots (ToF)

Fig. 6 shows the time-of-flight plots of the two scanned areas. Different buried layers have been individuated by

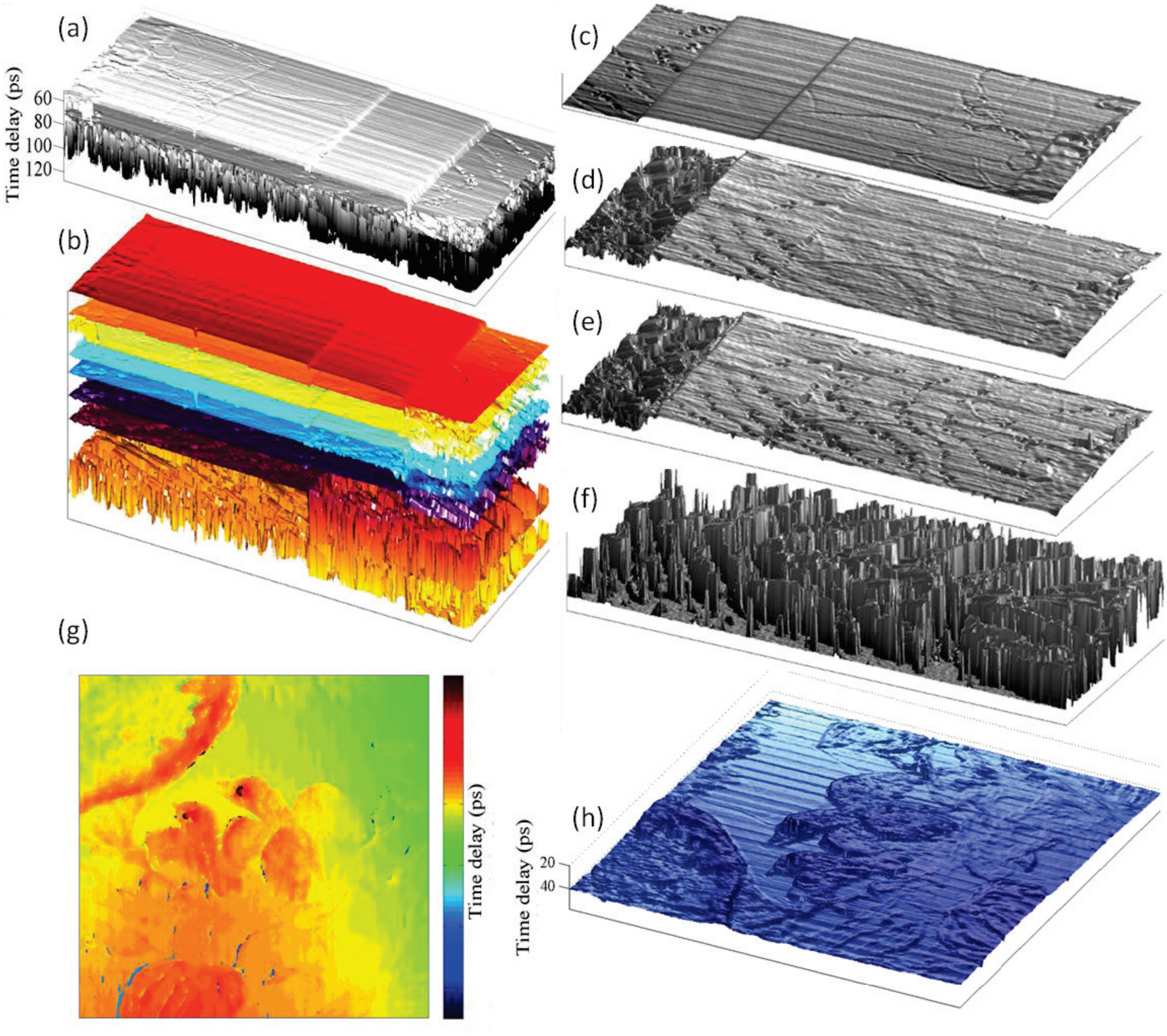

Fig. 6. (a) ToF image of the layers found for the area a2 of Fig. 2(a) in the real time-scale. (b) Arbitrary separation of the layers for a better comprehension. (c) ToF image of the surface. (d) Layer IV/layer III interface (interface I5 of Fig. 5(a). (e) Layer III/wood interface (interface I6 of Fig.5a). (f) Internal structure of the wood, plotted thanks to the optical inhomogeneities inside the panel due to the wood grain. (g) ToF image of the surface of the area a2 of Fig. 2(a), plan-type rendering. (h) ToF image of the surface of the area a2 of Fig. 2(a), 3D rendering.

plotting the maximum of the electric field for increasing time delay, in different time windows.

Fig. 6(a) shows the layers found for the area a2 indicated in Fig. 2(a) using the real time-scale of he reflected signal, while in Fig. 6(b) the layers have been arbitrary separated for a better comprehension.

Fig. 6(c) is the ToF image of the surface, Fig. 6(d) the layer IV/layer III interface (interface I5 of Fig. 5(a)), Fig. 6(e) the layer III/wood interface (interface I6 of Fig. 5(a)), Fig. 6(f) is the internal structure of the wood, plotted thanks to the optical inhomogeneities inside the panel due to the wood grain.

Fig. 6(g) and 6(h) represent the ToF images of the surface of the area a2 of Fig. 2(a), respectively a plan-type plot and a 3D plot.

Surface topology mapping of hidden layers allowed by the THz-TDI technique has a great importance in the field of artworks inspection for conservation or art-history purposes, enabling the detection of defects or characteristic features of the surface of buried layers. The possibility to visualize the internal structure of the wooden support allows the inspection of its integrity.

IV. CONCLUSION

Lacquerwares, ornamental objects decorated by multiple, composite layers of lacquers, are one of the most significant expression of Eastern Asian art. Scientific analysis of lacquers provides conservators with crucial information about composition and condition of the constituent layers, aiding in the development of appropriate conservation treatments. Here we have demonstrated that terahertz time-domain imaging (THz-TDI) in reflection geometry, being capable of highlight interfaces between layers in a stratigraphic buildup, can be a versatile technique for obtaining structural information about lacquered objects, providing not only 2-D images but also subsurface 3-D images and stratigraphic images (b-scans) contactless and non-invasively. We have investigated a European lacquerware replica by reflectometric THz imaging. The inspected lacquerware is a wooden panel covered by multiple complex layers of lacquers and plaster. Utilizing pulsed Terahertz Time-Domain Imaging (THz-TDI) in reflection mode, we observe non-invasively buried layers of the laquerware replica, including the internal structure of the wooden panel itself. With the same technique we have performed a surface material mapping. The visualization of material distribution has been enhanced through reflected THz composite RGB false color rendering, where RGB mapping allows distinction between different materials and textures on the surface of the lacquerware. To the best of our knowledge this is the first application of this color rendering technique to terahertz reflection images. The contrast between different textures is enabled by wavelength-dependent scattering from the surface, as well as differences in the composition of the surface layer.

ACKNOWLEDGMENT

The authors would like to thank Mikkel Scharff, head of Department and Associate Professor, at the Royal Danish Academy of Fine Art in Copenhagen (Denmark), for having provided the lacquerware replica investigated in this study and for the stimulating discussions we had togheter.

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