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Wall Painting Investigation by Means of Non-invasive Terahertz Time-Domain Imaging (THz-TDI): Inspection of Subsurface Structures Buried in Historical Plasters

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Abstract Characterization of subsurface features of wall paintings is important in conservation and technical art history as well as in building archaeology and architecture fields. In this study, an area of the apsidal wall painting of Nebbelunde Church (Rødby, Denmark) has been investigated by means of terahertz time-domain imaging (THz-TDI). Subsurface structures have been detected at different depths inside the lime-based plaster of the wall painting until approximately 1 cm from the surface. The surface morphology of the buried structures has been 3D imaged in detail, providing a substantial contribution in their characterization.

Keywords Terahertz · Wall paintings · Paintings · Cultural heritage · Imaging

1 Introduction

1.1 Potential of Terahertz Imaging for Wall Painting Investigation

To the best of our knowledge, the first interest in building material characterization by means of terahertz radiation was addressed to a typical office environment, for obtaining realistic model for propagation channels in terahertz communication systems (2005 [1]), approximately 10 years after the first terahertz time-domain imaging system was realized [2]. Simultaneously with the development of studies on the characterization of building materials for indoor wireless communication systems [3], GHz and THz frequencies started to be exploited for building material investigation within the context of non-destructive testing and civil structure damage detection. Imaging of concrete with microwaves (6–10 GHz) [4] and with terahertz imaging systems [5, 6] proved the ability to image metallic, dielectric objects and cracks inside concrete and building block unit samples. Systematic studies on the application of terahertz

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spectroscopy and imaging to construction, ground terrain, and building materials are still developing in the recent years [7, 8]. In the meanwhile, in 2006, the first attempt to investigate artworks by means of terahertz imaging systems was made [9], thus calling attention to the application of this technology to art investigation. Being the crossing point between paintings and building materials, the application of terahertz time-domain systems to the investigation of wall paintings followed rapidly [10]. The first attempt in the world to image an ancient real wall painting fragment was made in 2010 [11]. A number of case studies about terahertz imaging of wall paintings took place between 2013 and 2014, including mock-up samples [12–14] and real wall paintings of different ages (Neolithic [15], Paleolithic [16], ancient Roman [17], medieval [18]).

Within this research frame, this paper will show the use of terahertz time-domain imaging (THz-TDI) to image subsurface features of different natures until approximately 1 cm depth over an extended area of a real medieval wall painting (the apsidal wall painting of Nebbelunde Church, Rødby, Lolland, Denmark, 54.709079° N, 11.385661° E). By combining customized signal processing and advanced 3D visual rendering, the system provided a substantial contribution in substructure characterization and identification. At the same time, this study is a contribution to broaden the casuistry on the application of THz-TDI to wall painting investigation.

1.2 Subsurface Imaging of Wall Paintings: the Conservation Issue

The long-term preservation of mural paintings is linked not only to the preservation status of the surface pictorial layers but also to the state of the building substrate on which they are applied. A detailed knowledge of the internal structure supporting a wall painting may be considered the key to a successful restoration project. For preservation purposes, localization of cracks and detached areas, as well as identification of regions where filling or consolidation is needed, is as important as localization of buried or concealed structures inside the plaster. The presence of degrading, deforming, or moving components inside plaster layers can affect their integrity and consequently also affect the condition of the painting layers themselves. Furthermore, brick examination is at the basis of *mensiochronology*, while the analysis of stratigraphic units constituting walls and their reciprocal relationship is the fundament of relative chronological analysis of buildings and decorative surfaces, such as wall paintings. Accordingly, the characterization of subsurface features of wall paintings and position, geometry, and appearance of internal structural elements is essential for understanding building history and its construction phases and thus important in conservation and technical art history as well as in building archaeology and other architecture areas.

There are a large number of well-developed non-destructive methods for the investigation of wall painting subsurface features, including infrared reflectography, thermography, ground-penetrating radar, acoustic impact method (sounding), ultrasonic pulse velocity method, and ultrasonic pulse-echo method [19]. Advanced non-destructive testing (NDT) methods such as radar and ultrasonic and sonic methods are mainly suited for detection and characterization of large inhomogeneities at depths larger than 2 to 10 cm [20]. Thermography is mainly used for testing the near-surface region to a depth of up to 10 cm [21], while IR reflectography is a suitable method for detecting features, underdrawings, or engravings sketched by the artist on the preparation layer immediately behind the paint layers [22].

THz-TDI has recently proven its utility as a means of investigating wall paintings [10–18], panel paintings [23, 24], and easel paintings [25], and in reflection configuration, it has the

prospect of being a complementary non-invasive technique for subsurface feature investigation of historical plasters. Due to the extremely short electromagnetic pulses being emitted from a THz-TDI system, time-of-flight measurements are feasible, thereby allowing for 3D reconstruction of the subsurface structures. The technique may provide subsurface information to a depth of a few centimeters, at depths greater than IR reflectography, but lesser than thermal imaging. This intermediate depth range is of course important to explore since many classes of damage originate from defects which are close to the surface, such as delamination of plaster, surface and subsurface cracks, voids, spalling, soiling, efflorescence, and microbiological attack.

### 1.3 Terahertz Time-Domain Imaging in Reflection Geometry

Terahertz time-domain imaging (or terahertz pulsed imaging) is a non-invasive, coherent optical imaging technique that can acquire inherently 3D 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 (approximately 3–0.03 mm in wavelength or wavenumbers in the range 3–300 cm^{-1}) [26]. THz-TDI systems typically have a usable frequency range of approximately 0.1–3 THz (wavelength range 3–0.1 mm, wavenumber range 3.3–100 cm^{-1}), with the exact frequency range available for spectral measurements depending on specific key instrumental parameters, such as duration and power of the femtosecond laser pulse, composition of the semiconductor material, and design of the emission and detection antennae [27].

The attractive property of terahertz radiation for imaging applications is its ability to penetrate optically opaque and non-conducting materials. In a reflection geometry, the sign and amplitude of the back-reflected THz pulse can be analyzed for reflections originating from the various interfaces present between the sample layers (Fig. 1b), with a reflection amplitude determined by the terahertz refractive index mismatch at each interface, $r_{i,i+1} = (n_i - n_{i+1})/(n_i + n_{i+1})$. In general, this occurs when there is a structural or material change in the sample under investigation, which can result in strong reflections (echoes) of the incident beam even from buried layers. The temporal spacing between the reflections is proportional to the optical thickness of the layers (Fig. 1). Therefore, the time scale provides information about the penetrated depth of the signal that is reflected at each lateral position (x,y). In the majority of the systems in use, images are obtained by simple raster scanning of a terahertz beam across an object over the two lateral dimensions (x,y). The recorded quantity at each spatial coordinate is the electrical field strength of the electromagnetic pulse as a function of time (Fig. 1a).

The conversion of pulses from time to frequency domain by means of Fourier transformation could in principle allow substance identification through chemical mapping, owing to changes in the spectral content of the reflected THz pulses, given that many solids exhibit characteristic spectral features in the 10 GHz–4 THz frequency range. Although THz imaging has the potential to reveal information about the spatial distribution of the spectroscopic data, it is important to realize that there are many practical limitations to the capability of chemical mapping:

- Firstly, even if both phase (or timing) and amplitude information can be obtained, enabling the investigation of both morphological and chemical changes, the structural information and the chemical information are convolved in a single reflected pulse and can only be separated if one is known a priori.
- Secondly, THz images show a reduced frequency resolution compared with purely spectroscopic investigations mainly due to limited temporal windows of the signals [28]. It can be

Fig. 1 **a** Terahertz waveform; Δt_1 and Δt_2 indicate temporal spacing between the first/second and second/third reflected pulses, respectively. **b** Schematic representation of the incident terahertz beam $E_0(\omega)$ and the back-reflected signals $E_r(\omega)$, $E_{r'}(\omega)$, $E_{r''}(\omega)$, for a sample composed of two homogeneous layers in air investigated in reflection configuration at normal incidence; t_{ij} and r_{ij} indicate complex Fresnel field transmission and reflection coefficients for the different media: *1* (air), *2* (homogeneous layer), and *3* (homogeneous layer); n_i refractive indices of the three different media; d_1 and d_2 layer thickness; *I1*, *I2*, and *I3* the first, second, and third interface, respectively
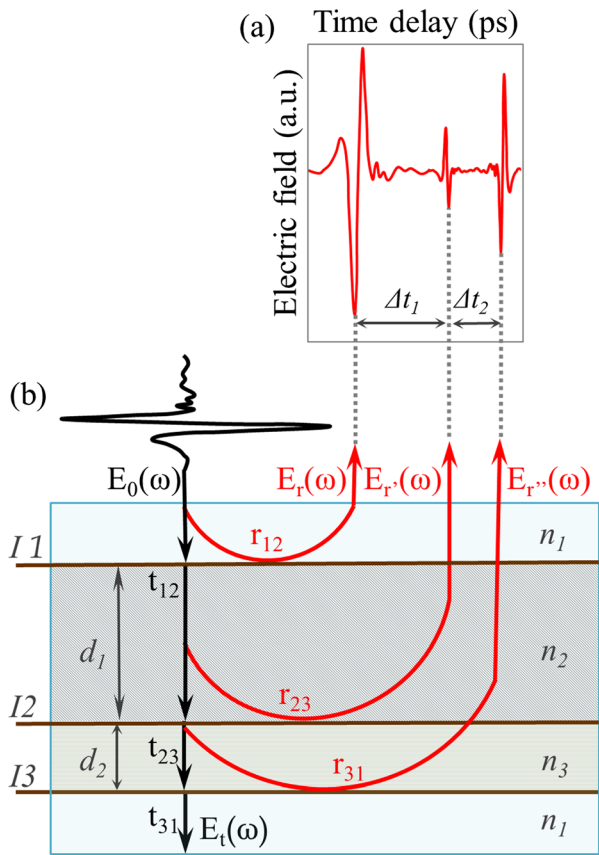
argued that the short duration of the pulse can be used either for depth information or spectral information. For a layer separation Δd, echoes will be separated by a time $\Delta t = 2n\Delta d/c$. This time defines the available time window for spectroscopy; thus, the highest possible undisturbed frequency resolution available for spectroscopic analysis of the reflected signal from the specific interface will be $\Delta\nu = 1/\Delta t = c/(2n\Delta d)$. Thus, for any situation where depth resolution and spectroscopic resolution are required simultaneously, the general uncertainty relation

$$\Delta\nu \cdot \Delta d \geq \frac{c}{2n} \tag{1}$$

must hold. As an example, for layers with a refractive index of 1.5, separated by 0.25 mm, the best frequency resolution available will be 0.4 THz, clearly limiting the capabilities for spectral recognition in layered structures.

- Thirdly, scattering of electromagnetic radiation in the THz range is considerable since the spatial scale of refractive index fluctuations—due to variations in either the surface roughness or internal structure of materials—can become comparable to the wavelength of the THz signal.

This issue is particularly relevant in relation to investigation of wall paintings, since THz-TDI measurements in reflection geometry are strongly influenced by the scattering of THz waves caused by irregularity, grain-like and uneven structures typically found in such

paintings. Recently, an image processing technique for the visualization of buried layers through uneven wall painting surfaces has been reported and the method is in progress [15].

Images recorded with THz-TDI can be plotted utilizing different parameters. In the time domain, images can be reconstructed from measured data using the already mentioned pulse delay with regard to a reference pulse (time-of-flight or ToF). Plan-type images (C-scans) can be displayed using different characteristics of the temporal amplitude $E(t)$ of a pulse, such as its maximum, minimum, or peak-to-peak values, the pulse area or power over a specific time interval (time windowing), and the centroid- or weighted return time of the pulse [29]. An additional time-domain parameter is the full width at half maximum (FWHM) of the pulse, which is also affected by the refractive index (especially if the medium is strongly dispersive) and frequency-dependent absorption coefficient [28]. In the frequency domain, images can be displayed by using the spectral amplitude $E(\omega)$ at a specific frequency or the amplitude or power integrated over a specific frequency range. Further parameters which have been used in frequency domain are the reflectance $R(\omega)$ and the corresponding reflection density, defined as the logarithm (base 10) of the reciprocal of the reflectance [30].

The lateral resolution (or spatial resolution) is restricted by the beam waist diameter of the focused beam, which is in turn limited by diffraction effects to approximately the wavelength of THz radiation utilized (Abbes criterium). In the frequency range from approximately 0.1 to 4 THz, the ideal lateral resolution ranges from 3 mm to 75 μm. 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 $L_C = c/(n\pi\Delta\nu)$, where $\Delta\nu$ is the FWHM spectral bandwidth and c is the speed of light in the intervening medium, with refractive index n. For a 0.5-THz FWHM spectrum in an environment with a refractive index of 1.5, the ideal depth resolution is thus approximately 65 μm. In practice, effects such as dispersion and scattering will reduce the depth resolution. Even if the depth resolution is physically limited by the spectral bandwidth of the terahertz radiation, different techniques have recently been used to improve the depth resolution [31, 32].

2 Instrumentation and Analytical Methods

THz-TDI was performed with a portable Picometrix T-Ray 4000 device, consisting of a femtosecond fiber laser coupled with 5-m-long umbilical cords to a photoconductive transceiver head mounted on an XY-scanning stage and a rapid-scanning pump-probe system for generating and detecting short pulses of electromagnetic radiation at terahertz frequencies. The frequency range covered is approximately 0.1–2 THz, and the signal-to-noise ratio (SNR) is about 80 dB at 0.5 THz. The relevant area has been scanned using a 320-ps measurement window, with a temporal resolution of 0.078 ps. The system has a fundamental data acquisition rate of 100 scans/s, and we employed a scan velocity of 12.5 pixel/s (eight full temporal waveforms averaged per pixel) in a reflection configuration at normal incidence. The raster scanning was performed with a lateral resolution of 400 μm, comparable to the optical resolution of the system (300 μm) at 1 THz, which is the highest frequency considered here. The optical resolution was determined independently by a knife-edge scan of the THz beam profile [33].

We have examined a region (approximately 18×14.6 cm, corresponding to 457×371 pixels) of the apsidal wall painting of Nebbelunde Church (Rødby, Lolland, Denmark). The investigated medieval wall painting (Fig. 2a) is of Gothic style and depicts Saint Anne, sitting on a bench with the infant Mary and holding a book on her left hand and the Christ in glory on the right. The pictorial technique used is the traditional medieval Danish one, which typically employs copper-based green pigment, carbon black, and earths [34]. After the Protestant Reformation (the sixteenth and seventeenth centuries), the wall painting was covered by a thick layer of limewash. Despite the past uncovering intervention, some lime-based mortar remains are still present on the surface.

3 Results and Discussion

3.1 Results and Discussion of Surface Inspection

Figure 2b shows the visible image of the investigated area. The value of the electric field recorded at each spatial coordinate (x,y) of the scanned areas has been used to generate both frequency and time-domain parametric terahertz images by a custom-written script for the computational program Matlab.

Figure 2c shows the grayscale plan-type terahertz reflection image, based on the peak-to-peak value of the reflected THz signal. Despite the characteristic high reflectivity of carbon-based pigment in the terahertz range [35], the outlines of the figures, painted with carbon black, are not always visible in the terahertz image due to strong scattering of the reflected
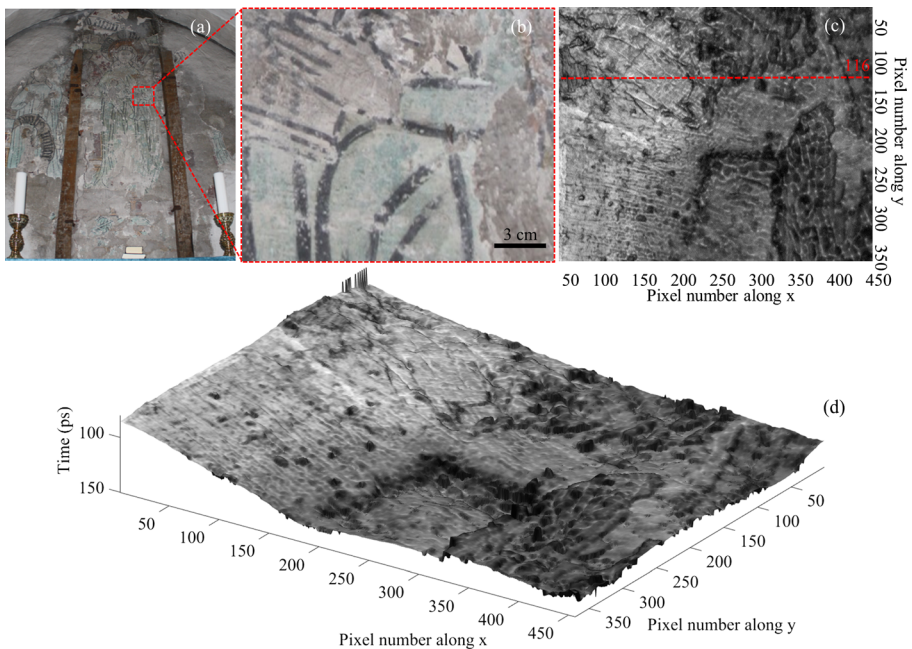

Fig. 2 a Apsidal wall painting in Nebbelunde Church, Rødby, Lolland, Denmark. b The relevant detail of the painting. c THz reflection image of the detail. The red dashed line indicates the scan line used for the B-scans shown in Fig. 3. d THz ToF image of the surface

signal by the irregular and uneven surface, which cause unquantified signal losses due to the imperfect coupling between the detector and the reflected beam. Regardless of this, it is possible to identify the carbon black distribution in some painting details, including the outlines of Mary's fingers (especially the index), the normal outlines which define the bottom of the book (particularly the one of the open page), and, even if pale, the two curved outlines defining the draperies of the dress that falls to her knees. The reflection image clearly localizes the incisions made on the wet lime-based plaster by a blunt point as preparatory drawing for the outlines of Mary's fingers (specially marked for the little finger) before the pigment application, which do not always overlap with the final painted outlines. This proves that the artist has changed his mind as to the composition during the process of painting (*pentimenti*). All the surface scratches appear highlighted, with an effect similar to that of grazing visible light. The lower, middle part of Fig. 2c shows a dark "Γ" shape mark that has no counterpart in the visible image.

3D ToF plot of the surface (Fig. 2d) reveals that this mark is related to an abrupt shallow depression of the wall at that point. The difference in return time of the signal between the depression and the surrounding area (5–10 ps) indicates a depth of approximately 0.75–1.5 mm of the depression. The uncovered regions of the wall painting are characterized by a smooth appearance, whereas the regions still covered by limewash appear much more uneven in the THz image. This correlates well with the visual appearance of the area.

3.2 Results and Discussion of Subsurface Inspection

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

The B-scans reveal the presence of two major in-depth anomalies. Figure 3a shows the B scan image obtained by scanning the investigated area along the red scan line $y=116$ in Fig. 2c, and Fig. 3b is the associated waveform at position $(x,y)=(175,116)$. Figure 3c shows a
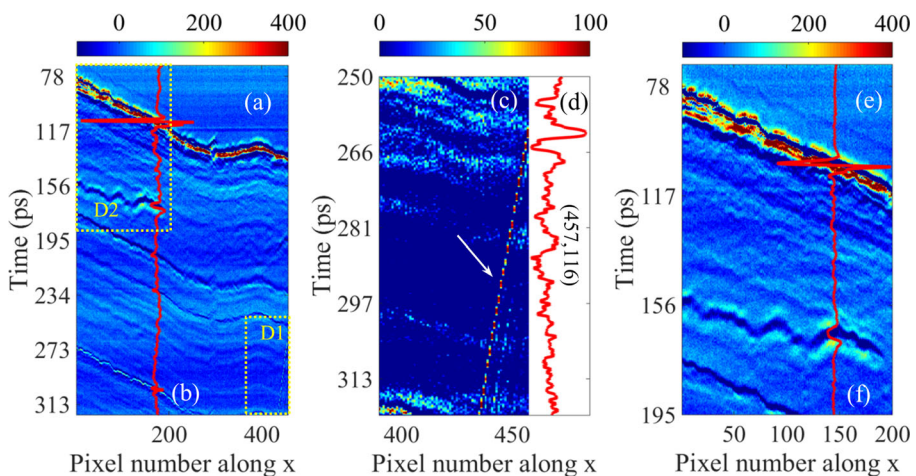

Fig. 3 *a* B-scan along the red line labeled 116 in Fig. 2c; the *yellow rectangles* D1 and D2 indicate the details plotted in *c* and *e*. *b* Waveform at position (175,116). *c* Detail D1 of the B-scan. *d* Waveform at position (457, 117). *e* Detail D2 of the B-scan. *f* Waveform at position (145,116)

zoom-in on the detail D1 where an in-depth interface is located. A waveform from the right edge of D1 is plotted in Fig. 3d. Within this temporal interval, from 150 ps after the arrival time of the main pulse until the limit of the scan window (320 ps), the anomalous interface is detected transverse to the plaster, highlighted by the white arrow in Fig. 3c. The systematic features that run across the B-scan image (Fig. 3a-c) parallel to the surface profile are in fact system echoes and not related to the internal structure of the wall. These features could, in principle, be suppressed by deconvolution techniques using a reference reflection signal from a metallic surface. However, these artifacts are easily recognizable and are discarded in the following discussion.

Assuming a reasonable average index of refraction $n=2$ [17] for the building material, the data show that the anomaly is located at about 1 cm below the surface ($\Delta x=c\Delta t/(2n)$). Closer inspection of the waveform in Fig. 3d shows that the reflected signal from the anomaly in D1 is of opposite sign than the reflection from the surface. Thus, the material related to the anomalous interface must be with a lower refractive index than the surrounding mortar [31].

ToF imaging again offers additional insight when interpreting the B-scan plots. Zooming in on the anomaly identified in Fig. 3 (region D1), we clearly visualize the relevant interface, located transversely at a steep angle into the wall. Detail of its surface morphology is shown in Fig. 4a. The extremely sharp and regular texture of the buried interface makes it most probably not an accidental air gap, but rather a building structure. The interface is found to be in close contact with a further interface that, given its characteristic surface texture (Fig. 4b), may be attributed to a further building element.

The other anomaly (D2 in Fig. 3a, e) is found at about 7–8 mm depth inside the plaster. This feature is present in all the raw B-scans for x pixel 1–250 and y pixel 50–224. Due the complex and the varying shape of this hidden feature, the recorded data are best visualized in a volume rendering, as shown in Fig. 5a-c. The locations of the anomaly is given by the time at which the maximum values of the temporal signals are located, where the data set was first processed by windowing to isolate the proper regions of interest before the volume rendering. The color map of the volume has been rescaled so that the internal feature visibility is maximized compared to the surroundings [36]. Figure 5d-g shows the location of this last anomaly found within the wall painting with respect to the surface. Here, the anomaly has been colored in yellow/orange tones to allow an easy differentiation from the surface, shown in green hues.

The exact nature of the detected anomaly is difficult to assign to a particular material. However, in contrast to the anomaly in D1, it seems that the polarity of the signal reflected from the anomaly D2 is the same as that reflected from the surface (Fig. 3e), indicating that this
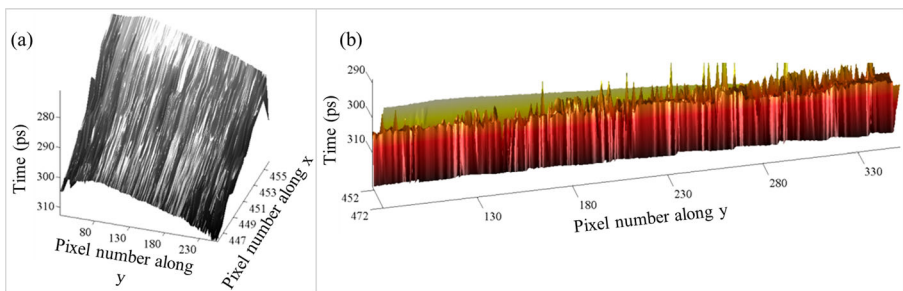

Fig. 4 a Detail of the ToF plot of the first buried anomalous interface found. b Detail of the ToF plot of the second buried interface found
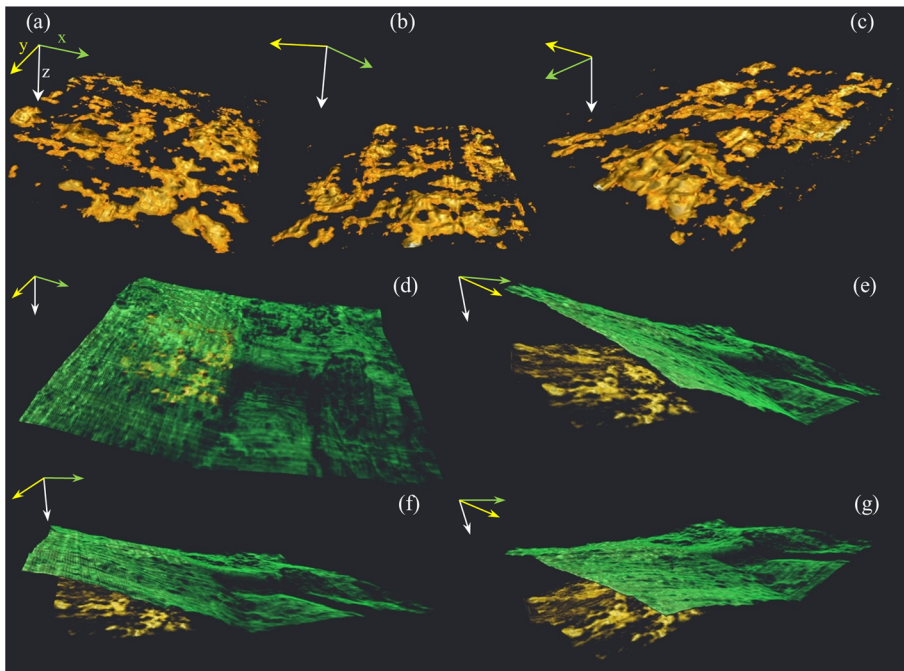
Fig. 5 *a–c* 3D representation from different viewing angles of the in-depth anomaly detected. *d–g* 3D representations from different viewing angles of the anomaly found at the location of the surface wall painting

anomaly has a higher refractive index than the material above it. This could indicate that the inclusion consists of a material with a higher density than the mortar [31]. An air gap or a material of low refractive index, even of sub-wavelength thickness, would lead to a reflected signal of opposite polarity compared to the surface reflection, as observed for the anomaly in region D1 of Fig. 3a-c [31].

4 Conclusion

The apsidal wall painting of Nebbelunde Church (Rødby, Lolland, Denmark) has been investigated by means of non-invasive terahertz time-domain imaging (THz-TDI). An anomalous interface characterized by high reflectivity values has been found at a depth of approximately 1 cm inside the lime-based plaster of the wall painting and imaged by B-scans and time-of-flight plots. The identified structure appeared to be in contact with a building element of different surface morphologies, which have been clearly 3D imaged. A second anomaly is found at about 7–8 mm in depth inside the mortar, as observed in the B-scans and visualized by 3D volumetric rendering. The sign of the reflected signal from the anomaly with respect to that from the surface indicates that the inclusion is a material with a higher refractive index than the surrounding mortar.

The interpretation of the building elements found in depth in plaster is rather difficult, and further scans over extended areas should be performed for certain identification of these elements. Despite this, the surface textures which characterize the structures found have been

3D imaged in detail by THz-TDI. This demonstrates how the proper THz data set processing combined with the use of advanced 3D visual rendering makes the THz-TDI technology interesting not only for identifying the presence of hidden subsurface inclusions in lime-based plaster of similar composition but also in understanding their surface texture. Given that the readability of the surface morphology of buried structures is an important step for attempting their identification, it follows that THz-TDI may provide information beneficial both for preservation and for typological analysis purposes.

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