

Review

Terahertz and Cultural Heritage Science: Examination of Art and Archaeology

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Abstract: Cultural Heritage scientists need methodologies to examine Art and Archaeology in order to understand artistic materials and techniques and devise better conservation procedures. This review discusses the most successful and promising applications of Terahertz (THz) technology in Cultural Heritage Science. THz is used in homeland security and for plenty of other industrial sectors and it presents a number of valuable features specifically for the investigation of Art and Archaeology: No radiation risk, low power, non-contact and reflection mode. Recent technical advancements are also making its application fast, mobile and relatively affordable creating a potential for its diffused implementation in museums. While THz is most promising for the investigation of multilayered art, such as paintings, it has been tested on a very large range of artifacts, from manuscripts to mummies and lacquered historical furniture.

Keywords: Terahertz; art conservation; conservation science; cultural heritage science

1. Introduction

Cultural Heritage Science (CHS) is a multidisciplinary academic science which encompasses sectors as Physics, Chemistry, Biology, and Engineering, and bridges them with Art conservation and Archaeology [1,2]. Cultural Heritage scientists examine works of art and archaeology by means of technical and scientific methodologies to understand when and how these artifacts were made, and, as well important, how are they to be preserved, what conservation treatment represents the best option and why.

A large number of imaging and spectroscopy techniques are commonly used by scientists involved in CHS. Among the most used imaging methods there are technical photography [3–7], ultraviolet photography [8], reflectance transformation imaging [9], infrared reflectography [10], multispectral imaging [11–13] and X-radiography [14]. Imaging methods are largely used since they do not need any sampling or treatment of the objects. On the other hand, analytical methods can be invasive and destructive. In many cases, however, sampling cultural artifacts is not permitted and, therefore, the application of non-invasive analytical techniques, such as reflectance spectroscopy [15], Raman spectroscopy [16], XRF spectroscopy [17] or neutron techniques [18,19] as well as mobile atomic force microscopy [20], is mandatory.

Terahertz (THz) radiation lies between the infrared and microwave regions (frequency range about 0.1–10 THz), Figure 1. With the advent of stable, sub-picosecond-pulse-duration lasers, researchers at Bell Laboratories in the mid-1980's explored the generation and detection of short transients of radiation that possessed a broad spectrum ranging from tens of gigahertz up to several THz [21]. These transients—which were generated by the laser excitation of DC-biased, fast-responding photoconductive gaps or non-centrosymmetric electro-optic crystals and detected using laser-triggered photoconductive sampling gates or electro-optic sampling—became known as

THz pulses [22]. Terahertz technology was used sparingly by few researchers for spectroscopy [23,24] and astronomy [25] until recent advances in instrumentation allow now to generate and detect terahertz with commercial equipment [26–28]. Research papers exploring THz applications are growing considerably and THz radiation is currently used in quality-control [29,30] and homeland-security [31,32] to screen for hidden weapons and explosives, since it can penetrate clothing. Its applications are also explored in medical-diagnostic [33,34] (mainly for cancer diagnosis [35]) and pharmaceuticals [36,37] for tablet inspection, process improvement and polymorph screening as well as to investigate coatings integrity [38].

This paper introduces THz technology and discusses its applications for Art and Archaeology. Its first test in this field dates back to 1998 when THz was investigated as a tool for dating wooden objects (dendrochronology) [39–41]. The same instrumental advancements that led the growing interest into this technology also generated research into a number of cultural heritage applications [42]. THz as a tool for Art diagnostics is still in its infancy because the technology is relatively less explored than other parts of the electro-magnetic spectrum, and many THz devices are relatively bulky and have not yet been optimized for use on-site in museums and in field sites. Yet the studies presented in this review point to a promising imaging and analytical method. THz waves can characterize many materials at different layers from the surface providing a visual understanding along the depth axis. THz data could yield more precise information about what is beneath the layer of an artifact – to better understand its materials and its current conservation condition.

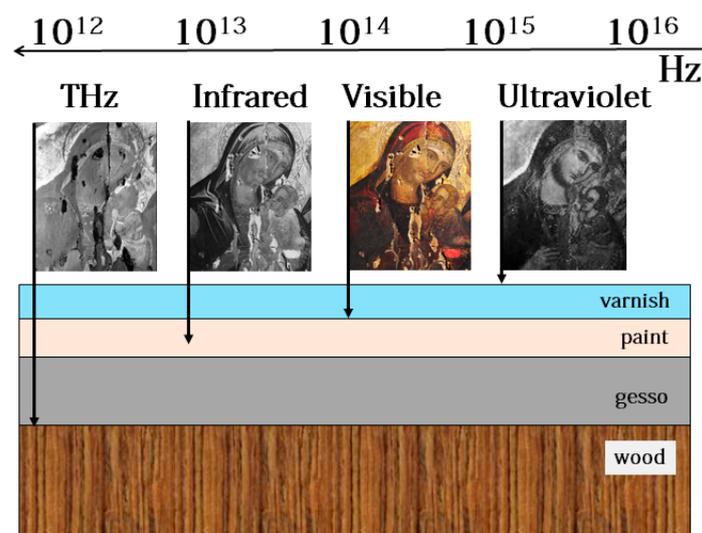


Figure 1. Terahertz radiation (10^{12} Hz) is often applied for the study of paintings. Terahertz (THz) and X-ray radiography can penetrate a panel painting deeper than other radiations traditionally used for paintings examination, such as infrared, but only THz allows for safe (non-ionizing) in deep 3D imaging of its layers.

2. THz Unique Features

THz is complementary to the other techniques commonly used for art examination and it provides different and unique information.

2.1. Optical Opaque Materials Become Transparent

THz radiation penetrates non-polar, non-metallic optically opaque materials (such as plastics, paper, painting materials and textiles) and it can be used for subsurface imaging since it penetrates much further into the sample than infrared techniques, commonly used in art examination.

2.2. Sensitive to Water

Unlike infrared radiation which induces molecular bending and stretching, THz excites collective motions, particularly in water molecules. THz radiation is so sensitive to water that it is used for biomedical studies of tissues whose water content is linked to important information. Humidity and water content are important information also in art conservation [43].

2.3. Safe

THz pulses have low energy and power and THz radiation is non-ionizing. In accordance with international safety guidelines moderate exposure to THz radiation is safe [44] and it doesn't heat the sample or cause damage [45]. The other nondestructive methods commonly used for sub-surface imaging in art and archaeology (X-ray, γ -ray, protons and neutrons) are ionizing and consequently their accessibility is limited.

3. Instrumentations

3.1. THz Range

Different wavelengths are appropriate for analyzing different types of cultural assets: art (notably easel paintings), for which THz radiation at the higher frequency range is preferred (0.5–13 THz), and historic architecture and archaeology, for which lower frequencies (0.01–0.5 THz) can be successfully applied for the analysis of thicker objects such as wall paintings and architectural structures.

3.2. Scanning

By focusing the terahertz beam with a short focal length lens, spatial resolution can be scaled down to tens of micrometers much better than sub-surface radars. THz waves lie between the infrared and microwave spectral regions, and therefore they combine the superior lateral resolution of infrared radiation (as compared with microwaves) with the higher penetration depth of electro-magnetic GHz-frequency fields (as compared with infrared). The high spatial resolution allows to scan the THz probes across the surface of the sample both in transmission and reflection mode. The probes are coupled with umbilical optical fiber cables, making it possible to scan the sample rather than move it and to quickly configure the scanner for transmission or reflection mode. The THz emitter usually delivers a picosecond duration THz pulse, allowing high-speed, scanned images to be easily produced acquiring a waveform at each position.

3.3. Time-Domain or Frequency-Domain

There are two main type of THz instruments and the use of each one depends on the specific goals of the examination [46], Figure 2. Time-domain based spectrometers (TDS) use short pulses (few picoseconds) to create an instantaneous wide frequency spectrum covering usually up to 3 THz. The time domain information is acquired with a Fast-Fourier transform analyzer and the absorption or reflection spectrum is then studied. This is a very powerful technique but is limited in its dynamic range due to the very low power provided by the ultra-short pulses. By gating the time domain signal, reflections from sub-surface discontinuities within a layered sample can be revealed, both in reflection and transmission mode.

Frequency-Domain is another type of analysis consisting in frequency measurements. In this case, a continuous or relatively short (ms) THz wave is sent and a frequency domain analyzer (THz receiver) is used to measure the reflected signal. The data is taken one frequency point at a time, each with an integration period of few ms. This method provides a much larger dynamic range but with a longer measurement time. Since the current systems are built using waveguides, they have limited bandwidth (around 100–150 GHz) and it is necessary to exchange waveguide bands in order to cover different frequency regions.

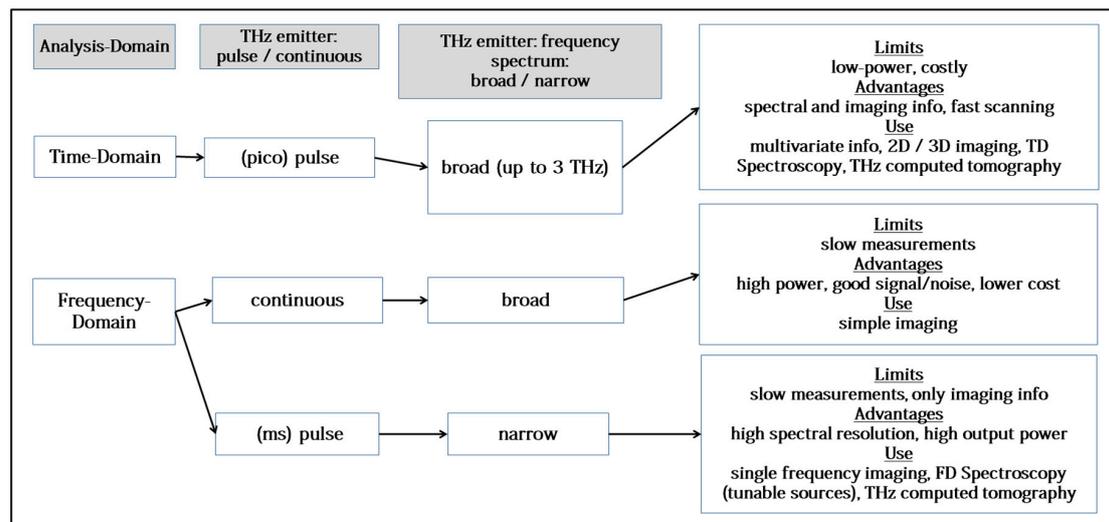


Figure 2. THz imaging and spectroscopy methods used in Cultural Heritage Science.

3.4. Pulse or Continuous Wave

Pulsed terahertz imaging is the most diffused method since it provides multivariate information on the sample. THz picoseconds pulses can be used in time-domain systems to generate images based on the pulse's peak amplitude or on its integration over a time window, or even its time delay [47–50]. On the other hand, continuous-wave systems are more affordable, have higher peak power and better image signal to noise. They are preferred for samples with simple subsurface structure.

3.5. Broad or Narrow Bandwidth

Most THz systems use broad bandwidth ultrafast THz pulses in Time-Domain for spectroscopy or imaging. A Fourier transformation of the time domain signal provides the reflected or transmitted spectrum of the signal, its amplitude or its phase. On the other hand, some THz scanners use narrow bandwidth THz emitters. They offer high spectral resolution and high output power but in order to collect spectra it is necessary to return the output frequency and therefore they are used mostly for single frequency imaging.

3.6. THz Spectroscopy

Spectral features in the terahertz region are due to molecular and inter-molecular interactions, weak bonds and phonon absorptions. Terahertz spectroscopy is implemented as time-domain (TDS) or frequency-domain (FDS). TDS systems use a broadband pulse signal and the transient terahertz electric field is sampled using an optical delay line while amplitude and phase spectra are delivered by Fourier transformation. In TDS it is possible to retrieve with a better signal-to-noise ratio the materials' complex index of refraction directly from the Fresnel equations rather than from the more complex Kramer-Kroenig relations. FDS is performed mostly by FTIR using broad-band and continuous sources and a two-beam interferometric spectrometer [51]. Regarding art materials there is an online THz spectra database by The National Institute of Information and Telecommunications Technology (NICT) [52,53].

THz spectroscopy of art materials is challenging for a number of reasons. Pigments' particle size fall within the terahertz wavelength range and Rayleigh and Mie scattering affect spectra significantly [54]. Also, most historical pigments have fillers and crystal polymorphs that impact the resulting THz spectra [55].

3.7. THz 2D and 3D Imaging

THz scanning can provide 2D imaging which is used for fast analysis of large surfaces but it is 3D imaging of sub-surface structures that makes THz so interesting for art examination. After a Time Domain Imaging scanning is acquired, it is possible to plot the amplitude at each point as a false color image [56,57]. Similarly to ultrasound scans, this THz image is called B-scan, when it shows just one scanning line, where the x-axis represents the position on the line and the y-axis indicates the time increment. The false color scale is related to the signal's amplitude. B-scan representation is used to show the sub-surface layers as a cross-section.

On the other hand, a THz image is called C-scan, if it shows an area, rather than only a scan line. The false color scale shows the intensity of the THz reflection at any desired subsurface depth (time delay). Signal processing such as apodisation [58], wavelet denoising [59] and deconvolution [60] is usually applied to enhance both B-scan and C-scan images.

3.8. THz Tomography

Terahertz computed tomography (THz-CT) features poor spatial resolution but in some cases it is preferred to X-ray since it doesn't affect thermoluminescence dating and it is sensitive to organic materials [61]. THz-CT uses mostly narrow bandwidth sources, multiple projection angles and back projection [62,63]. It is also possible to use broadband sources in time domain mode and to represent in the XY-plane for each pixel the time series (as depth Z) [64]. THz-CT imaging can also be performed with pulsed terahertz radiation in the broadband Time-Domain and even using one single projection it is possible to reconstruct the 3D internal structure of the sample thanks to the time-of-flight delay of the terahertz pulse [65].

4. THz for Art and Archaeology

4.1. Historical Documents

Often, historical documents on papyrus as well as on parchment and on paper cannot be read for a number of reasons. Sometimes the fragile sheets just cannot be separated because they are stuck together as a result of deterioration and damages. In other cases, the sheets have been reused as supports or covers for newer documents. There is considerable interest in reading this hidden information and a number of techniques have been tested to pursue this goal while preserving the documents. So far, X-ray computed tomography [66] has been the most successful method. Since THz-TD imaging was already evaluated for the inspection of postal envelopes [67], it was also tested with encouraging results for stacked papyrus layers written with carbon black ink [68]. Recently, even more successful results were obtained with a new and more sophisticated THz method called tomosynthesis [69] which was applied to image pencil writing on a stack of 50 paper sheets. THz tomography has been applied to resolve text on both sides of a single papyrus sheet [70,71]. THz was tested also for those cases where writing is obscured by stains and other inks in old parchment manuscripts [72] and it seems successful to characterize and evaluate conservation of iron gall inks [73] and parchment [74].

4.2. Panel Paintings

Diagnostics is preliminary to any conservation treatment in order to identify painting materials and technique, to detect previous restorations and additions as well as the nature and extent of alterations. Ultraviolet radiation is used for the examination of the varnish, infrared to reveal underdrawing and X-rays to study underpaint and the wooden support. THz has been successful to investigate all of these layers, even reaching the wood support, providing cross-section (B-scan) and top-view (C-scan) images of the artworks [75].

Gilded panel paintings are a particular kind of art developed in the early medieval age. They were made [76] applying a thin primer on the wooden panel support and then the gold background

on a layer of red bole. First attempts to apply THz for the examination of this technique dates back to 2009 when mock-up panels consisting of gold leaf and pigments on wood panel partially obscured by a gesso layer (chalk or gypsum preparation) were tested successfully [77].

THz is used mostly to locate the gilding leaves under the paint layer. Figure 3 shows a detail from a 14th century icon which was analyzed with both infrared imaging and THz [75]. In the THz-TDI image the square gilding leaves are clearly visible at the contours of the figures' faces while infrared reflectography (IRR) imaging cannot reveal them [78]. Before cleaning treatment, THz can be used for the inspection of the condition of the gilding leaves and of the decorative motives made with gold powder (shell gold), because THz can reveal gold even under a thick and dark varnish. Furthermore, the THz image of a panel painting at its different interfaces (wood, primer, paint) can reveal structural problems or paint's flaking. THz imaging of the wood grain reveals the cause of the cracks in the paint documented with infrared-RTI [78], Figure 3. Thanks to Terahertz's non-invasive cross-section imaging it is possible to evaluate the extent of cracks in the paint that are visible to the naked eye but whose depth and extent into the other sublayers cannot be determined otherwise [79]. It is also possible to visualize the traditional complex preparation, consisting in a canvas between two gesso layers [79].

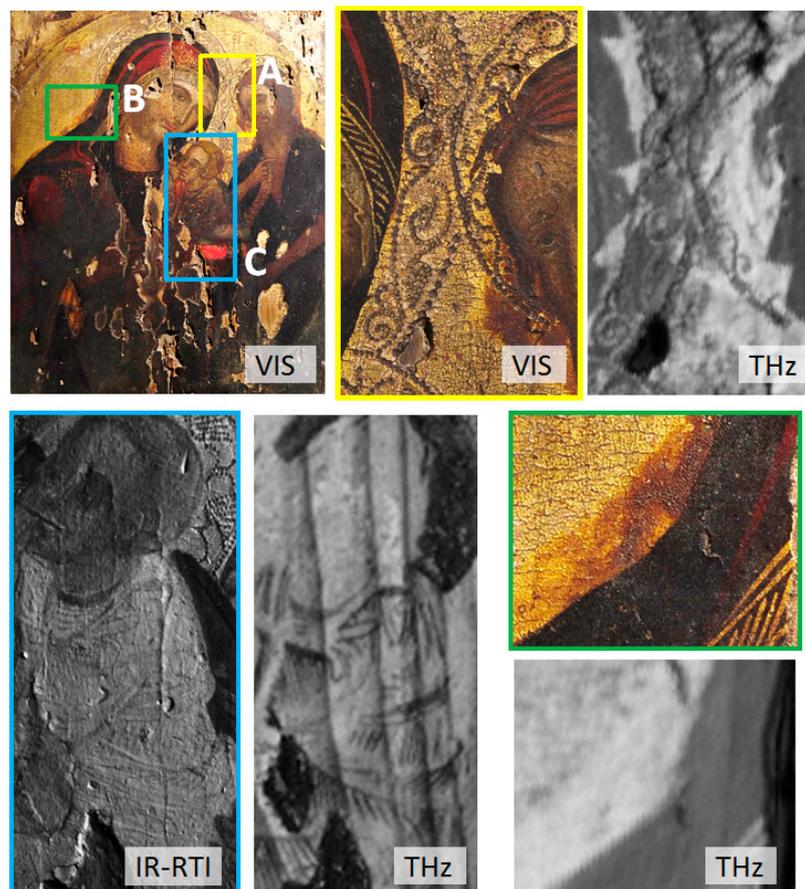


Figure 3. Virgin with Child and a Saint, 14th century icon (32 cm × 39 cm × 0.8 cm), Public Library in Taormina, Sicily. The icon was affected by extensive damages. Before its conservation treatment, it underwent a diagnostic campaign using THz and other imaging techniques (VIS (photography), IR-RTI (Infrared-Reflectance Transformation Imaging)). Each insert illustrates a specific application of THz for panel paintings examination: (A) gilding leaves' edges localization; (B) pre-cleaning visualization of gilding and gold decoration under dirt and varnish; (C) structural condition evaluation (interface wood-preparation). Courtesy of Danish Technical University [78].

4.3. Canvas Paintings

During the early Renaissance, painters started to prefer canvases to panels as supports for their paintings because they were much affordable and could be folded for more convenient transportation. Differently than wood panels, canvases are more transparent to THz and, indeed, the first test of THz for the inspection of paintings on canvas in 2006 [80] was performed with a transmission THz-TDS system. THz radiation has been used to detect underdrawings, changes (“pentimenti”), or previous compositions that are now hidden. While infrared imaging and X-ray radiography are the preferred methods for these kinds of cases, THz has the advantage of further penetration (compared to infrared radiation) and depth resolution not offered by standard X-radiography. The first test of THz for underdrawing observation dates back to 2008 when a THz-TDS in reflection mode could detect graphite pencil drawing under different pigments [81]. Underdrawing with umber was also tested in 2009 with a reflection THz-TDI system [82]. While graphite can be detected with infrared imaging, umber is transparent to the infrared and THz inspection is superior.

4.4. Wall Paintings

In 2008 it was demonstrated that THz imaging can reach through a depth of almost 1 cm into plaster [83]. This announcement started up a prolific research line dedicated to wall paintings that were whitewashed or even covered with a thicker layer of plaster [84]. Mobile and compact up-to-date THz-TDI scanners can be used for in-situ analysis of wall paintings. Their picosecond pulses are reflected back to the detector from discontinuities in the refractive index caused by cracks, covered paint layers or objects inserted in the plaster such as nails. In particular, THz scanning has been used to detect wall paintings whitewashed with lead white, which is very opaque to infrared imaging [84]. As for canvas paintings, THz can also map pigments on wall paintings [85] but the most important application on this specific kind of art is to evaluate the extent of cracks into the plaster [84].

4.5. Textiles

The interaction of THz with textiles has been studied extensively for homeland security applications [86,87]. This research has been instrumental to apply THz to study art and archaeology textile-wrapped objects such as mummies which are otherwise examined routinely with X-ray radiography or computed tomography systems in order to image their internal anatomical features [88,89]. THz adds the advantage of using non-ionizing radiation, portability and complementarity since in THz imaging contrast depends on the refractive index of the materials rather than the elemental density as in X-radiography. THz radiation does not have so much depth penetration as radiography but it provides high contrast images close to the surface useful to reveal the structure and condition of the bandages and objects placed within them [90]. On the other hand, THz systems are also used on small animal mummies and on mummified human body parts to study soft-tissues close to the surface [65].

4.6. Other Applications

THz studies on art and archaeology have been focused on historical documents, textiles and multilayered art such as paintings and lacquer furniture [91,92], Figure 4. There are other promising applications where THz has been tested even if less frequently. There is an increasing interest regarding THz application to detect and measure corrosion under paint for industrial applications [93–96]. This research could develop new THz methods for conservation of painted metal historical objects, such as furniture and sculptures.

THz spectroscopy has been tested on art made with plastics [97] since it is sensitive to their plasticizers and fillers, while THz 3D imaging can distinguish coatings and layers of degradation in historical plastics. There is interest into using THz imaging to obtain stratigraphic information regarding corrosion and encrustations layers of archaeological metals [98] and for historical stone

objects conservation [43]. THz tomography with specialized equipment has been used to examine clay Egyptian vessels [64].

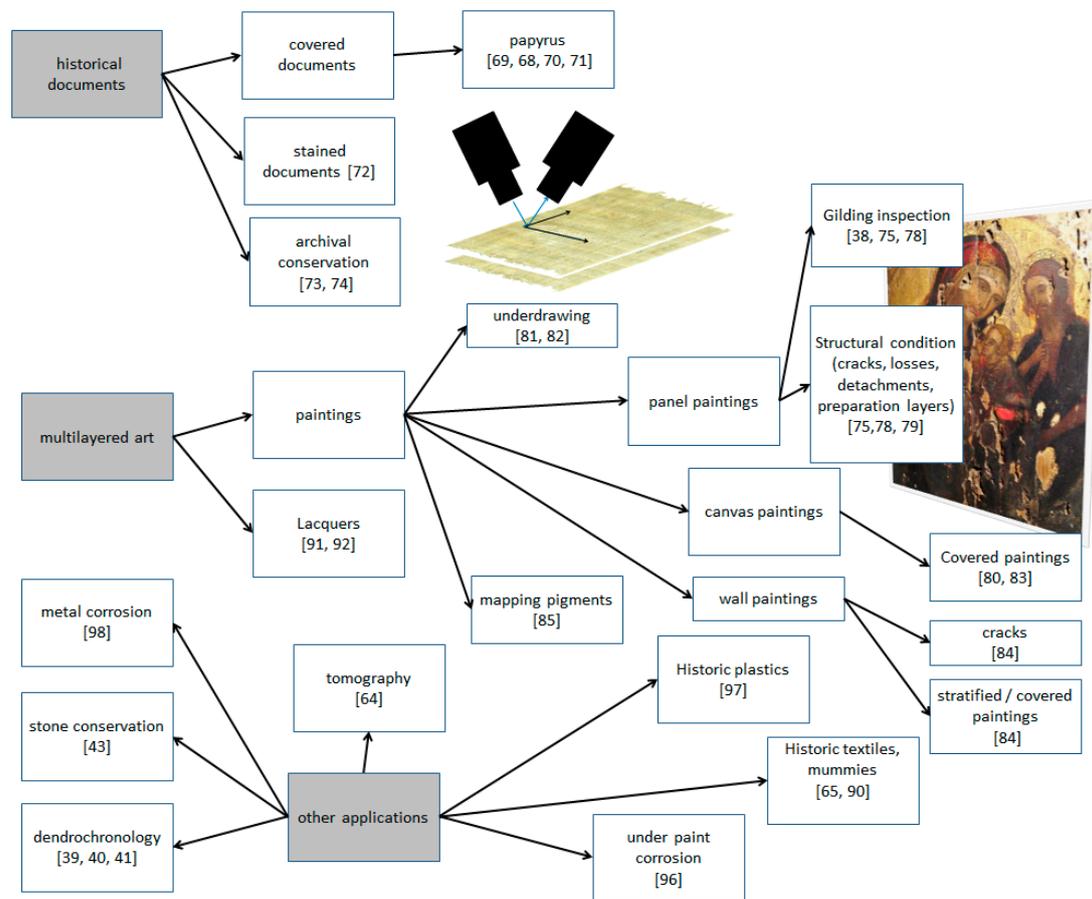


Figure 4. Applications of Terahertz in cultural heritage science.

5. Conclusions and Perspectives

THz technologies have experienced intense development, mainly because of their applications in homeland security, where depth profiling can be used to distinguish weapons and other dangerous items under clothing, and spectroscopy can be employed to sense explosive materials. Still a somewhat unexplored spectral range, Terahertz radiation offers a potentially important new tool for understanding art, searching for lost works, characterizing materials, authenticating artifacts, understanding decay processes, as well as improving conservation of paintings, murals and other cultural objects. Specifically to art investigations, use of the THz portion of the electromagnetic spectrum presents a number of valuable technical features: No radiation risk, THz radiation is non-ionizing and does not pose risk to health. Even if X-ray radiography and spectroscopy are used quite widely in art investigation, it is evident that the potential health and safety ramifications for the users of these methods would provide a strong incentive for the wide spread adoption of THz methods that would not require skilled personnel and specific facilities such as radiation-shielded rooms. Another advantage is that THz is a very low-power technique: The energy in a single THz pulse is typically less than the background radiation present, yet the signal-to-noise ratio can be very high due to the use of gated detection. Other advantages are that THz is non-contact, works in reflection mode, it is relatively fast and affordable. The historically decreasing costs of pulsed lasers and other optical components, and thus of the entire THz systems, creates a potential for broad use in museums. Furthermore, the relatively bulky THz experimental stations common to research

facilities have evolved into commercial, suitcase-sized instrumentation that can be easily brought into museums—for *in situ* examinations—and also easily moved from one conservation department to another. Tomography methods, such as CT-scan, have been used successfully for some exceptional initiatives, and even for investigations of mummies, but they cannot be used routinely for practical reasons (for instance, it is not likely that curators will habitually move art objects to external facilities like hospitals, due to limited budgets and security issues). THz tomography allows conservators to investigate thin layers, such those in a painting, as well as thick bulk samples, such as wood artifacts. This versatility is mandatory for the success of the method for practical budget reasons. If it finds applications in more than one conservation department, it has more chances to be implemented for the broadest benefit.

Recent technical developments are providing THz commercial equipment relatively small and lightweight. They are also becoming sturdy for transportation and less affected by environmental condition. Their portability is an extremely important feature for their actual use in the cultural heritage field.

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