Phase-Sensitive Reflective Imaging in the Terahertz and mm-Wave Regions Applied to Art Conservation

Non-destructive technologies, especially those based on THz sources, have been developed at the ENEA Research Centre in Frascati for monitoring applications in the Cultural Heritage Conservation field. Some of the most significant examples of research studies will be presented, with particular attention on those enlightening the specificity of the ENEA tools

Andrea Doria, Gian Piero Gallerano, Emilio Giovenale, Giovanni Messina, Ivan P. Spassovsky, Anne Cecile More, Alberto Petralia

Introduction

In the past few years, growing interest has been directed to find newer and newer applications for radiation in the Terahertz (THz) frequency range, including those in the field of analysis and conservation of cultural heritage [1].

THz radiation ranks in the electromagnetic spectrum between the Infrared (IR) and millimetres waves (mm-wave) and has the peculiarity of being a non-ionizing radiation capable of penetrating most plastics, cellulose and dielectric materials. The THz propagating through the material suffers of a minor scattering process if compared with infrared and visible radiation. In biological systems, absorption and reflection from water limit the penetration of THz radiation to the surface layers, but at the same time provide a powerful contrast mechanism that allows to detect the presence of biodegradation affecting the object. Despite the long wavelengths employed, Near Field Microscopy techniques can be used to reach high spatial resolution. Advanced THz technology for non-destructive analysis of art objects, including paintings, murals and sculptures has also good chances to become an imaging and analytical method for investigation. THz imaging and spectroscopy can indeed be used to characterize in a non-invasive way every layer and interface from the surface to the supporting material of a painting or mural. Accurate depth information can be provided by using short pulses and time-of-flight techniques or by using quasi CW radiation with phase-sensitive methods.

Andrea Doria, Gian Piero Gallerano, Emilio Giovenale, Giovanni Messina, Ivan P. Spassovsky
ENEA, Unità Tecnica Sviluppo di Applicazioni delle Radiazioni

Anne Cecile More, Alberto Petralia
ENEA Guests

The Imaging System

A versatile, reflective THz imaging system has been developed at ENEA-Frascati. The system is illuminated by a THz Compact Free Electron Laser operating in the frequency range from 90 to 150 GHz, with an output power of 1.5 kW in 4 μs pulses at a maximum pulse repetition rate of 10 Hz [2]. The imaging set-up is shown in Fig. 1 [3].

The FEL radiation is coupled first into a focusing cone followed by a circular to rectangular waveguide

transition, which matches the cone output to a series of two WR6 directional couplers. A waveguide probe is attached to the second directional coupler directing the FEL radiation to the sample under investigation.
The side outputs of the two directional couplers provide a reference signal (-20 dB) of the FEL radiation incident on the sample and the signal reflected by the sample (-16 dB) respectively. Both signals are detected by Schottky diodes operating at room temperature. A variable attenuator allows for the relative adjustment of the reference signal to the reflected signal within the response range of the Schottky diode during calibration. The sample under investigation is placed on top of a XY translational stage driven by piezo-motors with 50 mm travel range on both axes. To obtain an image, the sample is scanned at a maximum rate of 5 pixel/s with a maximum spatial resolution of about 0.2 mm, the reflected signal is normalized to the reference signal to compensate for any power fluctuation of the source during the scan. Collected data are stored in matrix form to be subsequently analyzed by an image processing software. The distance between the sample and the imaging probe along the z axis can be adjusted by means of a stepper-motor driven stage. It is worth pointing out that this imaging setup can be easily extended to higher frequencies up to about 1 THz by using a combination of both commercial and custom made waveguide components.

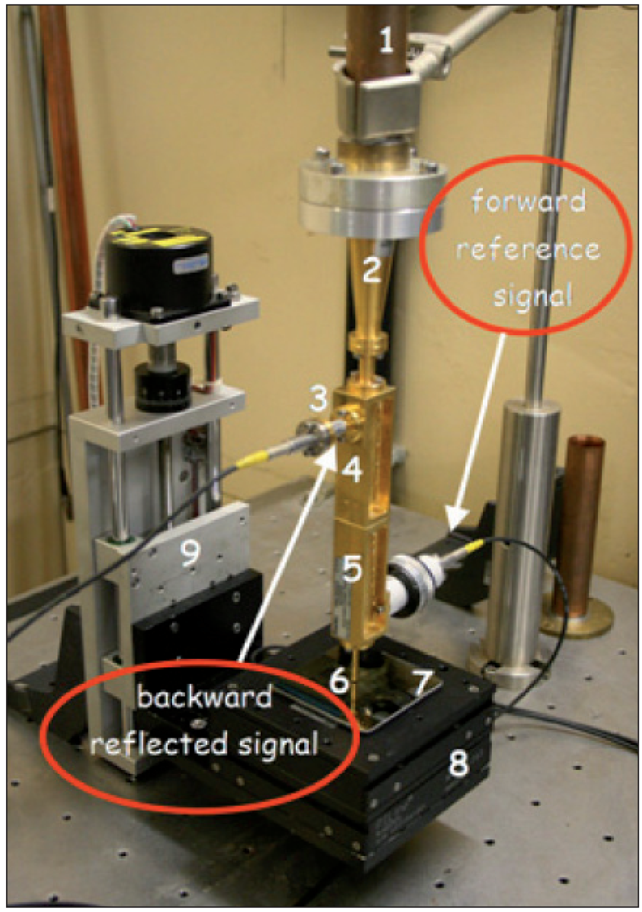

FIGURE 1 Photograph of the imaging setup: 1-Light-pipe; 2-Focusing cone; 3- Schottky diode; 4- 10 dB directional coupler; 5- 20 dB directional coupler;6- WR6 imaging probe; 7- Sample plane; 8- XY axes translational stage; 9-Z axis translational stage
Source: [3]

Experimental Results

Within the framework of a bilateral agreement, signed between the Italian and Japanese foreign offices, and named THz-Arte, we started a collaboration with Dr. Kaori Fukunaga from NICT (National Institute of Information and Communications Technology) about the application of THz radiation for the study of art work, in order to provide a tool for the conservation of cultural heritage. A first test measurement has been taken on a painting made with natural pigments and gold on a wood tablet, and partly covered by a 1.5 mm layer of whitening. This sample has been analysed in the visible (see Fig. 2 a), at THz in transmission using the broad band source at NICT (Fig. 2 b), and finally in reflection at Frascati with the ENEA Compact FEL source (Fig. 2 c). The whitening layer is perfectly transparent in the THz range and, due to the phase contrast control, in ENEA we obtained a clearer image right over the plaster cover: the whitening acted as an antireflection coating [4].
In order to verify the possibility to distinguish the pigments under the coverings, also the painting reported in Fig. 3 has been analysed. This is a tempera representing a Lady Mary's dress. Part of the detail of the dress has been covered by another kind of plaster, the *Gesso di Volterra.* The detail covered is now made

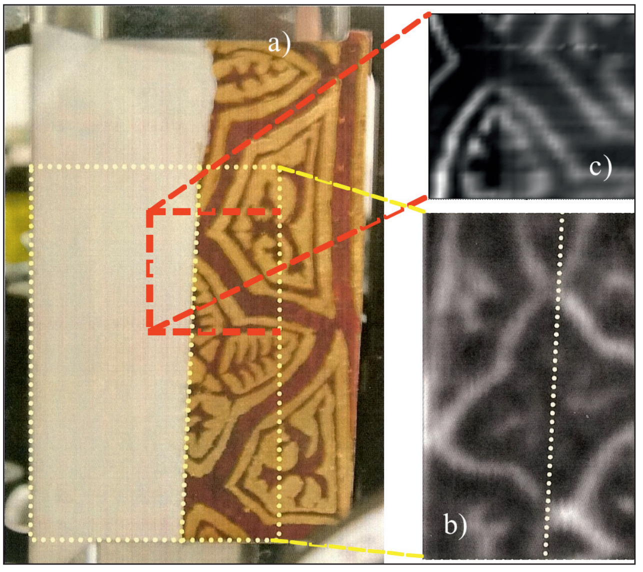

FIGURE 2 a) Visible image of a tempera painting partly covered by plaster; b) THz image in transmission @ 0.6-2.6 THz of the detail shown in the dotted area of the visible image (NICT Japan); c) ENEA THz image in reflection @ 0.15 THz of the detail shown in the dashed square of the visible image
Source: [4]

of two different pigments: indigo and cinnabar over gold. The THz image shows how the experimental apparatus is capable to distinguish not only the painting covered as being completely uncovered, but also the two different pigments demonstrating that they also have two different refractive indices at the ENEA FEL frequencies.

Another example is reported in Fig. 4 where the image represents a saint with a halo made of gold dotted; the painting has partly been covered with lampblack and white plaster. The resulting image demonstrates that the radiation can pass through these materials also.
The phase sensitivity of our imaging system has been exploited to distinguish the different refractive index of the different materials. The basic principle is based on the fact that the radiation, travelling along the waveguide of the probe (element 6 in Fig. 1), splits in two waves at the exit boundary; the transmitted part is attenuated by a factor and carries a phase term to which we must add a phase term related to the complex reflectivity of the sample. The reflected signal then interferes with the reflection from the open end of the waveguide, providing phase information, which can be used to enhance the image contrast and to monitor topological features of the sample surface. The amplitude of the maxima and minima interferences decreases as distance increases due to the radiation diffraction. For a metallic sample, the expected phase shift is π whereas for any other material the phase is a function of the complex refractive index. A proper mathematical model has been set up to match the experimental data with the free parameters. In Table 1 we have reported some results, obtained with the described method, about the optical characteristics of some materials used in cultural heritage and art conservation.
The phase-sensitivity method offers a powerful tool with which we can optimise the contrast among

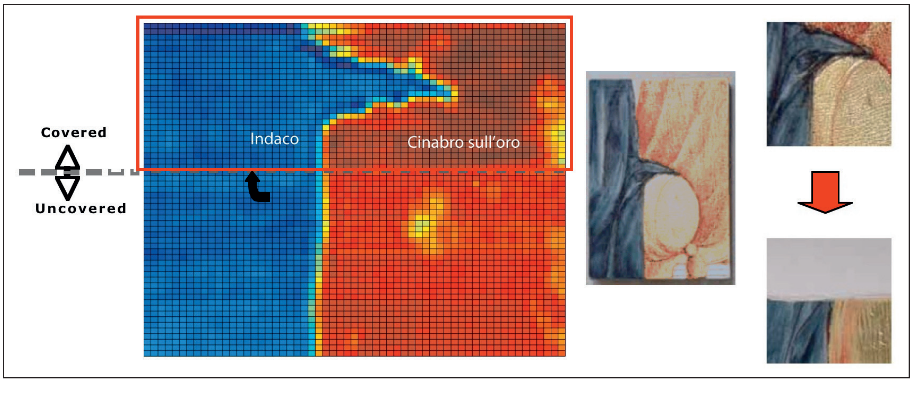

FIGURE 3 Tempera paint partially covered with plaster. Detail of the dress of the Virgin Mary. THz image and comparison with the visible image
Source: [4]

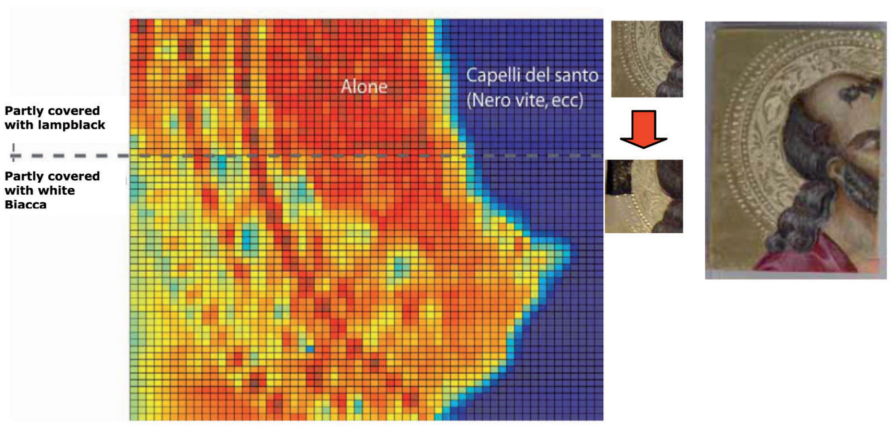

FIGURE 4 Tempera paint partially covered with carbon black and lead white. THz image and comparison with the visible image
Source: ENEA

TABLE 1 Optical parameters of some materials
Source: ENEA

	Re (n)	Im (n)	α [cm^{-1}]
Cyclo-Olefin Polymer (COP) base	1.04	0.81	25.01
Blue Cobalt Oil on COP	1.46	0.06	1.81
Blue Cobalt Acrylic on COP	1.30	0.26	8.06
Oil	1.15	0.36	11.36
Wax	1.32	0.28	8.35
Balm	1.42	0.11	3.10

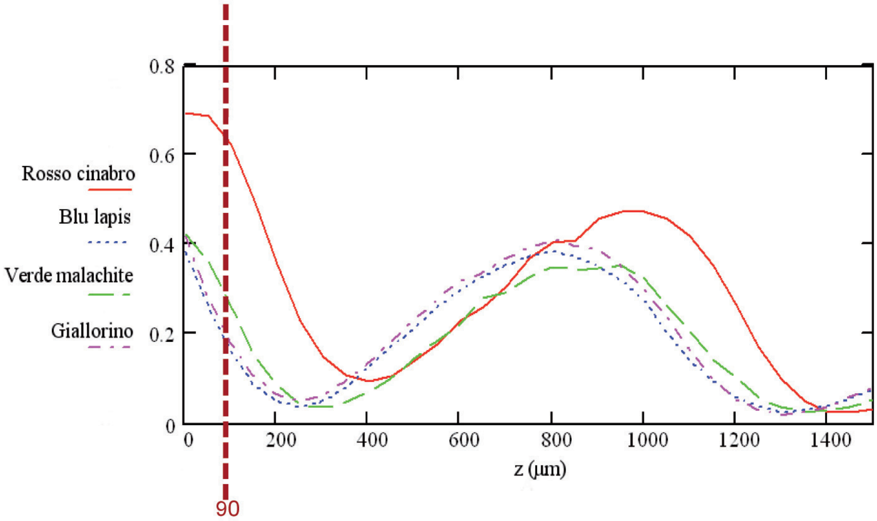

FIGURE 5 Phase scanning for different pigments. Highlighted in red is the distance for which the maximum contrast is obtained
Source: ENEA

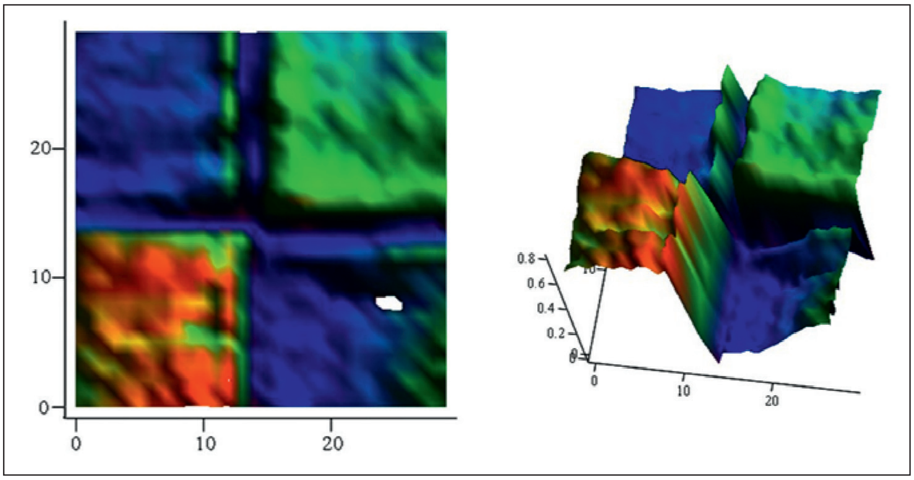

FIGURE 6 THz images of 4 pigments measured at the optimum distance obtained with the phase measurements
Source: ENEA

pigment responses in reflection. To proof the technique, a mosaic composed by four different pigments, was prepared (see Fig. 6); a phase profile scan, just moving the z-linear stage and thus changing the distance between the tip of the probe and the sample, has been performed for each pigment (see Fig. 5). A detailed analysis of the figure tells us that at a distance of 90 μm we have a maximum phase contrast for at least three pigments out of four. Setting the probe distance to that specific value, a 2D scan was performed.

A new activity recently started in Frascati on a complete series of samples provided by CISA3 (University of California, San Diego, USA) and prepared in 1983 at Editech, Florence (see Fig. 7). All samples reproduce typical material preparation for painting pigments, used for realising works of art over the centuries. The combination of 26 different pigments, painted with 5 different thicknesses over 3 different substrates, using 2 different binders, results in a total of 780 samples, covering most of the experimental situations [5].

Since THz radiation penetrates dielectrics, we expect to be able to identify the response from pigments, binders and substrates: by comparing the optical behaviour of the same pigments with different substrates and binders we will be able to determine the contribution of each component. Four different

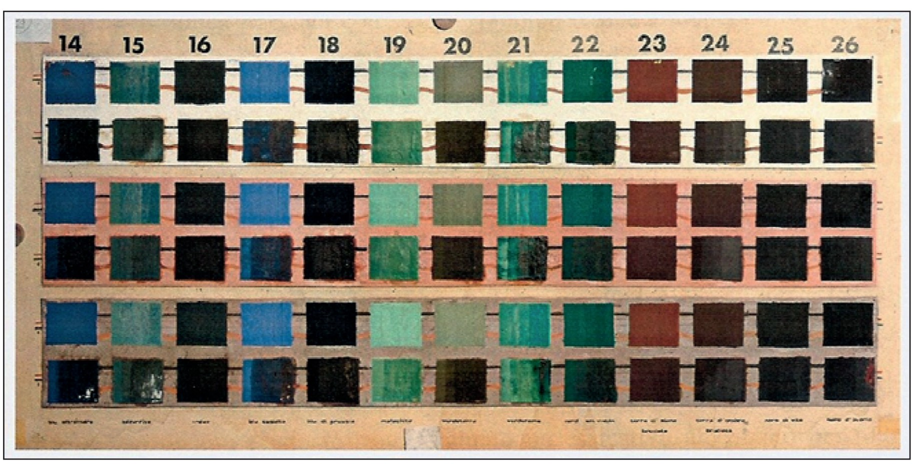

FIGURE 7 Panels with pigments prepared by Editech for THz measurements
Source: [5]

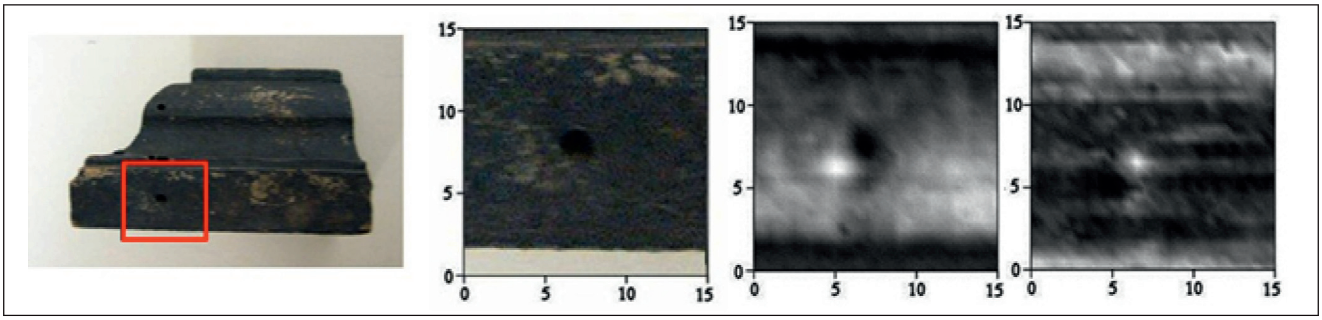

FIGURE 8 Fragment from a wooden altar dated 1774. From left: visible image, a magnifying of a particular and two THz images in reflection, measured at different tip-surface distances
Source: [6]

"wire patterns" were deposited between the substrate and the painted samples using the following materials: carbon, lead, silver and yellow ochre. It's important to verify if those materials can be detected under the painting, because these are the main components used for the preparatory drawings, usually underlying the final painting.
The three degree of freedom associated with the ENEA THz imaging system allow for tomographic imaging. This technique has been applied to detect damages induced by parasites, like worms, in wood samples. Reflectivity of wood is low, but it is sufficient to perform the measurements, if polarization of the Compact FEL radiation is chosen to be parallel to the wood fibres. The investigated samples are wooden objects from an altar dated 1774 of the Church S. Maria Maggiore, Piedimonte Matese (Caserta, Italy) [6]. The phase-contrast capabilities of the imaging system allows to perform a tomography scan, revealing internal properties of the sample and thus Fig. 8 reports visible and THz images, taken at different z-values, of the wood sample. It is quite evident how the hole changes its position in the image while changing the tip-sample distance, giving us evidence of how the tunnel develops beneath the surface.

Due to the capability of THz radiation to have a high contrast in reflection with respect to water content in samples, THz imaging techniques have been proposed to study the biodegradation of the mosaics in the "Villa del Casale" (Piazza Armerina, Italy). This will be a joint research activity between ENEA Frascati Reserch Centre and Centro Regionale per la Progettazione e il Restauro (CRPR) in Palermo. The reflection of the watery content of the biomaterials can be used to make a map over an area of the mosaic floor and to control its preservation state over time. A preliminary phase of the study will be carried out in laboratory to typify the material present in the mosaic works in order to plan the diagnostic operations on *in situ* mosaics at best and in a more targeted way. In particular, it is important to determine the transmission coefficients, reflectivity and the refractive index of the different materials, the characteristics of the different infiltrated microorganisms, in order to recognize them in the subsequent diagnostic interventions on the original mosaic. The second step concerns the diagnostic intervention *in situ* to map the infiltrate materials (such as algae, lichens, green patinas, etc…) under the mosaic "tesserae" without removing them and without deteriorating the mosaic themselves.

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